

1 **Regime-based Aerosol-Cloud Interactions from CALIPSO-MODIS and the Energy Exascale**
2 **Earth System Model version 2 (E3SMv2) over the Eastern North Atlantic**

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12 **Abstract.** This study investigates aerosol-cloud interactions in marine boundary layer (MBL) clouds
13 using an advanced deep-learning-driven synoptic-regime-based framework, combining satellite data
14 (CALIPSO, vertically resolved aerosol extinction and MODIS, cloud properties) with 1° nudged Energy
15 Exascale Earth System Model version 2 (E3SMv2) simulation over the Eastern North Atlantic (ENA;
16 ~10°×10°, 2006-2014). The E3SMv2 captures observed seasonal variations in cloud droplet number
17 concentrations (N_d) and liquid water path (LWP), though it systematically underestimates N_d . We then
18 partition ENA meteorology into four synoptic regimes (Pre-Trough, Post-Trough, Ridge, Trough) via a
19 deep-learning clustering of ERA5 reanalysis fields, enabling regime-dependent aerosol-cloud
20 interactions analyses. Both satellite and E3SMv2 exhibit an inverted-V LWP- N_d relationship. In Post-
21 Trough and Ridge regimes, the satellite shows stronger negative LWP- N_d sensitivities than in Pre-Trough
22 regime. The Trough regime displays a muted satellite LWP response. In comparison, the model predicts
23 more exaggerated LWP responses across regimes, with LWP increasing too quickly at low N_d and
24 decreasing more sharply at high N_d , especially in Pre-Trough and Trough regimes. These exaggerated
25 model LWP sensitivities may stem from uncertainties in representing drizzle processes, entrainment, and
26 turbulent mixing. As for N_d susceptibility to aerosols, N_d increases with MBL aerosol extinction in both
27 datasets, but the simulated aerosol-cloud interactions appear oversensitive to meteorological conditions.
28 Overall, E3SMv2 better captures aerosol effects under regimes that favor stratiform clouds (Post-Trough,
29 Ridge), but performance deteriorates for regimes with deeper, dynamically complex clouds (Trough),
30 highlighting the need for improved representations of those cloud processes in climate models.

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68 **1. Introduction**

69 Marine boundary layer (MBL) clouds play a pivotal role in regulating the Earth's energy budget
 70 due to their extensive coverage over the oceans and high albedo (Albrecht et al., 1995; Wood et al., 2015;
 71 Dong et al., 2023; Wall et al., 2023). Central to the quantification of the radiative impact of MBL cloud
 72 properties is to determine the sensitivity of clouds to the presence of aerosols. Aerosols can impact cloud
 73 microphysical properties, such as the cloud droplet number concentration (N_d) and droplet effective
 74 radius (r_e), and, consequently, alter cloud optical and macrophysical properties, including liquid water
 75 path (LWP) and cloud fraction (Twomey, 1977; Albrecht et al., 1989; Zheng et al., 2020; Dedrick et al.,
 76 2025). The interactions between aerosols and clouds, commonly termed aerosol-cloud interactions (ACI),
 77 contribute to one of the largest uncertainties in climate projections (IPCC, 2021). These uncertainties in
 78 the aerosol-induced cloud microphysical responses and the accompanying cloud adjustments stem from
 79 the inherent complexity of cloud microphysical processes (e.g., droplet activation, precipitation
 80 suppression, and entrainment-induced evaporation) and their interactions with dynamically evolving
 81 MBL, where aerosol perturbations can potentially contribute to either the brightening or darkening of
 82 clouds (Wall et al., 2022; Feingold et al., 2024).

83 Satellite remote sensing observations are essential in efforts to quantify the cloud adjustment to
 84 aerosol perturbations, by providing spatially extensive datasets. Numerous studies using satellite data
 85 have demonstrated a significant relationship and progressively advanced our understanding of cloud
 86 adjustments to aerosols (Bellouin et al., 2020; Diamond et al., 2020; Yuan et al., 2023; Feingold, et al.,
 87 2025; Goren et al., 2025). Observational evidence frequently shows that, at lower N_d conditions, with the
 88 increased aerosol concentration, cloud droplets become more numerous in smaller sizes. It leads to
 89 decrease in efficiency in colliding and coalescing into raindrops and suppress precipitation (Albrecht,
 90 1989). This suppression results in less cloud water being lost through rainfall and, consequently, an
 91 increase in LWP. Furthermore, the combined effects of entrainment-sedimentation feedback and
 92 precipitation-stabilization may lead to weaker entrainment drying in relatively clean clouds, further
 93 enhancing the LWP under lower N_d conditions (Bretherton et al., 2007; Wood, 2012; Possner et al., 2020).
 94 In contrast, at higher N_d levels, the increased abundance of small droplets expands the surface area
 95 available for evaporation at the cloud top (Gupta et al., 2021; Zhang et al., 2022; Zheng et al., 2022a).
 96 This enhancement in evaporation promotes cooling and intensifies localized turbulent mixing, which, in
 97 turn, facilitates the entrainment of dry air from above the cloud. The subsequent mixing further
 98 accelerates the evaporation of cloud droplets, reducing the overall liquid water content and decreasing

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104 LWP. However, distinguishing causality from correlations is a persistent challenge, as atmospheric
105 variability, retrieval biases, and sampling limitations can obscure true aerosol-induced effects (Arola et
106 al., 2022; Goren et al., 2023; Liu et al., 2024).

107 Parallel to observational advances, the modeling community has made significant progress in
108 simulating aerosol-cloud interactions within global climate models (GCMs). Recent GCM versions
109 incorporate more physically based cloud microphysics parameterizations, which enable the simulation
110 of precipitation suppression and enhanced entrainment responding to aerosol changes. For instance,
111 studies by Mülmenstädt et al. (2024a) show that some of the GCMs in the Coupled Model
112 Intercomparison Project Phase 6 (CMIP6), namely the US Department of Energy (DOE) Exascale Earth
113 System Model version 1 (E3SMv1; Golaz et al., 2019) and others, can capture the inverted V-shaped
114 relationship between LWP and N_d , as often observed from satellite retrievals, although discrepancies
115 persist regarding the causal interpretation of these relationships. Tang et al. (2024) highlights that even
116 when E3SM version 2 (E3SMv2; Golaz et al., 2022) simulates the overall cloud macrostructure, the
117 microphysical responses to aerosol perturbations are still subject to systematic uncertainties related to
118 precipitation processes and turbulence-microphysics interactions. Collectively, these studies illustrate
119 both the strides made and challenges remained in representing aerosol-cloud interactions in large-scale
120 models. While some GCMs may successfully replicate observed negative LWP- N_d relationships under
121 the present-day conditions, they struggle to accurately simulate the turbulence, entrainment, and
122 precipitation feedback, that govern cloud adjustments to aerosol changes (Mülmenstädt et al., 2024b).
123 Assessing model performance in simulating cloud microphysical responses to aerosol perturbations using
124 observation is a crucial step toward improving the process-level understanding.

125 Accurate assessment of aerosol effects on cloud microphysics faces several hurdles.
126 Methodological inconsistencies in sampling, aerosol proxies, and comparison metrics limit direct
127 observation-model comparisons. Studies that use vertical aerosol extinction profiles rather than column,
128 integrated aerosol optical depth report stronger links between aerosols and cloud microphysics,
129 highlighting the need for refined observational strategies (Painemal et al., 2019, 2020). Furthermore,
130 satellite retrieval errors, vertical mismatch between aerosol and cloud layer, updraft variability, and
131 precipitation effects, can bias the estimates of cloud sensitivity to aerosol perturbations (Quaas et al.,
132 2020; Gryspeerdt et al., 2022; Jia et al., 2022; Alexandri et al., 2024). Uncertainties in cloud adjustment
133 processes, particularly the balance between precipitation suppression and entrainment driven evaporation,
134 remain a persistent source of discrepancy between observational inferences and model simulations,
135 (Mülmenstädt et al. 2024b, Zhang and Feingold 2023). These challenges are compounded by the complex,
136 multiscale nature of ACIs, where small-scale processes interact nonlinearly with larger-scale

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184 meteorological drivers. Moreover, synoptic systems organize boundary-layer clouds on multi-day
185 timescales and strongly modulate aerosol-cloud-precipitation pathways (Mechem et al., 2018; Lee et al.,
186 2025). Therefore, quantifying the untangled aerosol-cloud sensitivities require conditioning on the
187 synoptic environment. For example, Zhang et al. (2022) found that the relationship between LWP and
188 N_d is not only sensitive to aerosol loading but also modulated by the underlying meteorological conditions.
189 And McCoy et al. (2020) used a cyclone compositing approach to demonstrate that aerosol-cloud
190 interactions (e.g., the sign of LWP change with N_d) can differ inside vs. outside midlatitude cyclones.
191 These considerations motivate our use of an objective synoptic-regime classification to control
192 meteorology when evaluating the synoptic-regime-dependent ACI.

193 The Eastern North Atlantic (ENA) region is uniquely advantageous for advancing our
194 understanding of ACIs in MBL clouds (Wood et al., 2015; Tian et al., 2025). Located at the confluence
195 of subtropical and midlatitude air masses, the ENA is characterized by diverse meteorological conditions
196 and cloud regimes. This region frequently experiences well-organized stratocumulus cloud decks and
197 other MBL cloud types, which are sensitive to both local and long-range transported aerosols (Wang et
198 al., 2020; Wang et al., 2022). Observations document distinct aerosol and cloud properties, and the
199 relatively pristine marine background punctuated by episodic aerosol events helps separate aerosol-driven
200 cloud adjustments from meteorological variability (Zheng et al., 2022b; Varble et al. 2023; Christensen
201 et al. 2024; Qiu et al., 2024). Moreover, the ENA has been extensively sampled, with long-term
202 observational data collected at multiple spatiotemporal resolutions from various platforms including the
203 DOE’s Atmospheric Radiation Measurement (ARM) research facility (Wood et al., 2015), and several
204 satellite remote sensing products such as those from the Cloud Aerosol Lidar and Infrared Pathfinder
205 Satellite Observations (CALIPSO; Winker et al., 2009, 2010), and the MOderate Resolution Imaging
206 Spectroradiometer (MODIS) on board Terra and Aqua (Barnes et al., 1998; Platnick et al., 2003; King
207 et al., 2013). These comprehensive observational datasets make the ENA an ideal testbed for evaluating
208 model aerosol-cloud interactions.

209 In this study, we employ a novel regime-based evaluation framework that combines the recently
210 developed CALIPSO-derived vertical-resolved aerosol extinction profiles and MODIS-derived cloud
211 properties with simulations from the DOE E3SM version 2 (E3SMv2). By applying a clustered-
212 meteorology-regime-based analysis over the ENA, we aim to isolate the impact of aerosol perturbations
213 on cloud microphysical properties by regime and seek to provide a robust observational and modeling
214 framework for quantitatively assessing aerosol-induced cloud adjustments. This approach is not only
215 intended for reconciling discrepancies between satellite observations and model simulations but also for
216 informing about potential model uncertainties in ACIs in connection with specific meteorological

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regimes. The data and methods used in this study are introduced in Section 2. The seasonality of aerosols,
clouds, and their interactions are introduced in Section 3. And more importantly, Section 4 presents the
meteorological-regime-based analysis of the aerosol-cloud interaction and cloud adjustments. Section 5
summarizes conclusions with discussion, and outlines future work.

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2. Data and Method

2.1 Satellite retrievals of aerosols and clouds

In this study, aerosol extinction coefficient (σ_{EXT}) is a research product derived from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) on the CALIPSO. These retrievals are produced at a 1 km along-track resolution using the Fernald–Klett iterative approach, constrained by an independent CALIOP-based aerosol optical depth (AOD), and are limited to cloud-free pixels. The retrieved profiles have been evaluated against airborne High Spectral Resolution Lidar (HSRL) measurements during the Caribbean 2010 field campaign and show good agreement. The detailed aerosol retrieval methodology and product evaluation are described in Painemal et al. (2019) and Li et al. (2022).

Cloud properties, including the liquid water path (LWP) and the cloud droplet number concentration (N_d), are obtained from MODIS Aqua at 1 km resolution using Clouds and Earth’s Radiant Energy System (CERES) Edition 4.0 algorithms (Minnis et al., 2021). The MODIS-retrieved LWP is estimated to have uncertainties of approximately 10–15% when compared with ARM surface-based observations (Xi et al., 2014; Painemal et al., 2016). The N_d is retrieved using the adiabatic formulation (Painemal and Zuidema, 2011; Grosvenor et al., 2018):

$$N_d = \Gamma^{1/2} \frac{10}{4\pi\rho_W^{1/2}k} \frac{\tau^{1/2}}{r_e^{5/2}},$$

Where the cloud droplet effective radius (r_e) and cloud optical depth (τ) are estimated from MODIS 3.79 μm and 0.64 μm bands, respectively (Painemal et al., 2013, 2020). Γ denotes the adiabatic lapse rate of condensation (Albrecht et al., 1990), which is calculated from the cloud-top temperature and pressure derived from MODIS (Painemal et al., 2020). ρ_W is the water density and k represents the ratio between the cloud droplet volume mean radius and the effective radius, assumed to be constant at 0.8 (Martin et al., 1994).

261 Although the relative errors in N_d retrieval can be significant at the pixel scale (Grosvenor et al.,
 262 2018), previous studies have shown that the N_d compares well with measurements from 11 aircraft
 263 campaigns, demonstrating a decent correlation when sampling the marine stratocumulus clouds, with r^2
 264 values of 0.5~0.8 (Gryspeerd et al., 2022). Therefore, to minimize known retrieval uncertainties, we
 265 focus on low-level liquid clouds where satellite N_d shows the strongest aircraft agreement and typical
 266 normalized root mean squared deviation of ~30-50 % (Gryspeerd et al., 2022). Moreover, the aggregated
 267 collocation method significantly reduces the MODIS Aqua N_d bias (Painemal et al., 2020), resulting in a
 268 relationship between aerosol and cloud properties less affected by artifacts. Note that to avoid diurnal
 269 variations in aerosol-cloud relationships, we fix the sampling to the Aqua local-afternoon overpass and
 270 do not merge with Terra morning orbits, while extending the collocation and quality-control framework
 271 to Terra is left for future work.

272 To collocate aerosol and cloud retrievals from the two satellite datasets, the following matching
 273 method is employed. For each 1-km CALIOP aerosol pixel, five 1-km MODIS pixels are selected on
 274 each side of the CALIPSO track (thus 10 MODIS pixels in total). The CALIOP retrievals are first
 275 averaged to produce a 5 km along-track resolution product, and the MODIS cloud retrievals are
 276 aggregated into four 5 km $\times$ 5 km grids (two grids east and two west of the CALIPSO track). These
 277 datasets are then further averaged over approximately 25 km along-track segments, ensuring that the
 278 aerosol and cloud data are matched at similar spatial scales. Hence, the clear-sky aerosol extinction
 279 profiles can be collocated in the vicinity of the clouds, enabling the ‘simultaneous’ assessment of aerosol-
 280 cloud interaction from the satellite data around 1 p.m. local time over ENA region. In addition, the
 281 CloudSat Cloud Profiling Radar (CPR) is used to determine precipitation status from the satellite. The
 282 drizzle condition is defined as the ratio of pixels with maximum radar reflectivity between -15 and -7
 283 dBZ to the total number of pixels (19) within the 25 km satellite collocated segment, while the thresholds
 284 for light rain and rain conditions are defined as maximum radar reflectivity greater than -7 dBZ but less
 285 than zero dBZ, and greater than 0 dBZ, respectively. For a detailed description of the data-matching
 286 strategy, please refer to Painemal et al. (2020) and Li et al. (2025).

287 To simplify terminology, all aerosol and cloud properties retrieved from the different satellite
 288 products are hereafter referred to collectively as “satellite” data.

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311 **2.2 E3SM simulations**

312 E3SMv2 is a fully coupled Earth System model (Golaz et al., 2022). Its atmospheric component,
313 EAMv2, closely follows its predecessor EAMv1, as described in Rasch et al. (2019) and Xie et al. (2018),
314 with only minor updates to its physical parameterizations. EAMv2 employs a spectral element dynamical
315 core with approximately 110 km horizontal resolution and 72 vertical layers. The radiation and aerosol
316 treatments in EAMv2 follow the Rapid Radiative Transfer Model (RRTM; Mlawer et al., 1997) and the
317 four-mode version of the Modal Aerosol Module (MAM4; Liu et al., 2016; H. Wang et al. 2020),
318 respectively. Turbulence, shallow convection, and cloud macrophysics are handled by the Cloud Layers
319 Unified by Binormals (CLUBB) scheme (Larson, 2017; Golaz et al., 2022), while stratiform cloud
320 microphysics is simulated by the Morrison-Gottelman (MG2) scheme (Gottelman and Morrison, 2015).
321 Deep convection is represented by the Zhang and McFarlane Scheme (Zhang and McFarlane, 1995), as
322 in EAMv1, but with a revised convective triggering function in EAMv2 that improves the simulation of
323 precipitation and its diurnal cycle (Xie et al. 2019).

324 In this study, EAMv2 was run at standard resolution (~110 km) with the meteorology nudged to
325 ERA5. The model was nudged toward the ERA5 zonal (U) and meridional (V) wind and temperature
326 fields using a relaxation time of 6 h. This nudging reduces errors in the simulated large-scale state,
327 facilitating the examination of aerosols and clouds. The nudged simulations reduce errors in simulated
328 meteorological conditions, facilitating the examination of aerosol and clouds. Hourly outputs are
329 available from 2006 to 2014 over the ~10°×10° ENA domain (33.5–43.5°N, 23–33°W), comprising 54
330 model columns. The σ_{EXT} profile is directly outputted from the model. The MBL cloud samples are
331 defined below 680 hPa to better match the satellite observations, and a cloud fraction threshold greater
332 than 5% is used to determine a valid MBL cloud layer as in Kang et al. (2024). To further compare with
333 the MODIS-retrieved cloud-top height (CTH), CTH is inferred by the diagnosed inversion height in
334 E3SM. The inversion height is determined where $\left(\frac{\partial \theta_l}{\partial z}\right) \left(\frac{\partial RH}{\partial z}\right)$ is minimized, with the θ_l denoting liquid-
335 water potential temperature and RH denoting relative humidity derived from the model outputs. Given
336 the coarse vertical resolution of E3SM near the cloud top, (~200-300 m), this approach accounts for strong
337 thermodynamic inversions and the effects of entraining dry air from the free troposphere (Erfani et al.,
338 2022). The cloud-base height (CBH) is similarly identified using the 5% cloud fraction threshold. The
339 in-cloud N_d is obtained from grid-box-averaged cloud liquid number, weighted by cloud fraction at each

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vertical level. Lastly, the cloud LWP is computed by integrating the in-cloud liquid water content (LWC) between cloud-top (CTH) and cloud-base (CBH) levels: $LWP = \int_{CBH}^{CTH} LWC \, dz$.

2.3 Clustering Model

Clustering meteorological patterns allows us to systematically characterize and categorize the diverse atmospheric conditions that modulate aerosol-cloud interactions in the ENA region. By identifying distinct meteorological regimes, we can, to some extent, isolate the aerosol-driven microphysical responses from the meteorological variability. In this study, we adapted an advanced deep learning-based clustering model proposed by Faruque et al. (2023), which features the architecture of the convolutional neural networks (CNN), and long short-term memory (LSTM) layers combined with a Deep Embedded Clustering (DEC) framework. This hybrid CNN-LSTM-DEC model was designed to capture complex spatiotemporal dependencies in meteorological data, overcoming limitations associated with conventional clustering methods that often treat spatial and temporal features separately (Zheng et al., 2025).

The clustering model was applied to ERA5 reanalysis data over a $\sim 10^\circ \times 10^\circ$ domain ($33.5\text{--}43.5^\circ\text{N}$, $23\text{--}33^\circ\text{W}$) in the ENA region for 2006–2014, with data at 13:00 LT (1 p.m. local time) daily to better match the time of satellite records. Input variables included 500 hPa geopotential height (Z500), mean sea level pressure (SLP), and the 10-meter u and v wind components. The CNN-LSTM-DEC architecture employs a sequence-to-sequence autoencoder with an encoder comprising four convolutional blocks (with filter sizes of 64, 128, 256, and 512), each followed by max-pooling to distill spatial features. An LSTM layer with 512 units captures temporal dependencies, and a dense layer with 256 units (using ReLU activation) defines the compressed latent space. A key aspect of our study was the fine-tuning of model hyperparameters through the grid search technique, which enabled us to systematically optimize clustering performance. The optimal configuration utilized a Stochastic Gradient Descent (SGD) optimizer with a learning rate of 0.01, momentum of 0.95, and a batch size of 32.

We first pretrained the CNN-LSTM autoencoder using a reconstruction loss. Then, the latent features extracted by the encoder were clustered using K-means, with which clustering into four groups yielded the highest silhouette score of 0.267, compared to scores of 0.257, 0.178, and 0.167 for five, six, and seven clusters, respectively. Hence, the determination of final cluster numbers (four, in this study) is

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375 based on a combination of silhouette score analysis (i.e., measures of cluster cohesion and separation for
376 different cluster numbers) and the sensitivity of aerosol and cloud property distinctions to the chosen
377 number of clusters.

378 To further refine the clustering assignments, we then ran DEC with that fixed cluster number of
379 four, as determined with K-means optimization. DEC was initialized by the K-means centroids and
380 optimized the KL-divergence clustering loss (between soft assignments and a sharpened target
381 distribution) with periodic centroid updates, which increased the silhouette score to 0.358, indicating
382 enhanced cluster cohesion and separation. This two-step clustering process significantly reduced intra-
383 cluster variability while accentuating differences between clusters, as suggested by Faruque et al. (2023).
384 Their work also demonstrated that integrating both CNN and LSTM layers produces more robust latent
385 representations, which are the reduced-dimensional encoding of the input that captures its most
386 significant attributes, compared to CNN-only models or traditional approaches such as K-means and self-
387 organizing maps. Furthermore, Zheng et al. (2025) showed that including additional meteorological
388 variables, notably the 10-meter wind components, improved the model’s ability to distinguish subtle
389 synoptic regimes over the ENA region compared to studies that considered Z500 only (e.g., Mechem et
390 al., 2018). Overall, the refined CNN-LSTM-DEC model demonstrates a marked improvement in
391 clustering performance over traditional methods for analyzing large-scale meteorological phenomena.

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396 **3. Aerosol and cloud properties from satellite and E3SMv2**
 397 **3.1 Seasonal distribution of cloud properties**

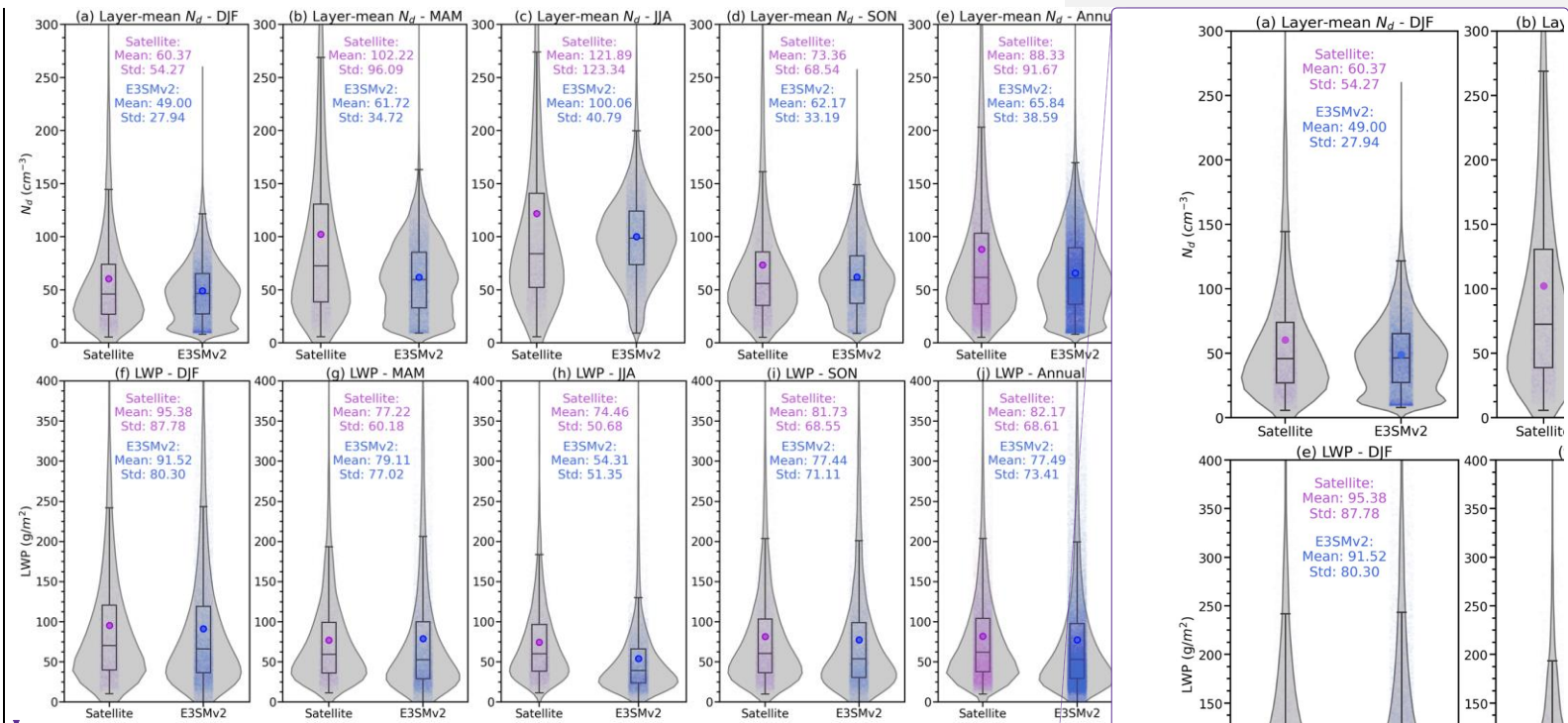


Figure 1. Violin plots of cloud droplet number concentration (N_d , top panels) and cloud liquid water path (LWP, bottom panels) from satellite retrievals (purple) and E3SMv2 simulations (blue), during winter, DJF (a, f), spring, MAM (b, g), summer, JJA (c, h), fall, SON (d, i), and Annual (e, j). The mean value is indicated by the color-coded dot. The smoothed shape of each violin shows the Gaussian kernel density estimate (KDE). From top to bottom within each violin, the box plot lines represent the third quartile (Q3, 75th percentile), median (Q2, 50th percentile), and first quartile (Q1, 25th percentile), respectively. The upper whisker extends to $Q3 + 1.5 \times \text{IQR}$ (interquartile range), and the lower whisker extends to $Q1 - 1.5 \times \text{IQR}$.

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Figure 1 illustrates the seasonal variations in the N_d and LWP for low-level clouds over the ENA, from satellite (MODIS) retrievals and E3SMv2. Annual means are $88.33 \pm 91.67 \text{ cm}^{-3}$ and $82.17 \pm 68.61 \text{ g m}^{-2}$ for satellite N_d and LWP, and $65.84 \pm 38.59 \text{ cm}^{-3}$ and $77.49 \pm 73.41 \text{ g m}^{-2}$ for E3SMv2 (Fig. 1e and 1j). Satellite-derived N_d exhibits a pronounced annual cycle, with the highest mean values during summer (JJA, 121.89 cm^{-3}) and spring (MAM, 102.22 cm^{-3}), followed by fall (SON, 73.36 cm^{-3}), and the lowest during winter (DJF, 60.37 cm^{-3}). E3SMv2 reproduces the N_d trend with a JJA peak 100.06 cm^{-3} and a DJF minimum 49.00 cm^{-3} but underestimates N_d in every season (Fig. 1a–1d), with the largest low biases in MAM 40.5 cm^{-3} and JJA 21.83 cm^{-3} . Satellite observations display broader distributions with higher variability, especially during MAM (96.09 cm^{-3}) and JJA (123.34 cm^{-3}). In contrast, the E3SMv2 exhibits generally narrower N_d distributions, with distinct peaks at low concentrations in all seasons except JJA. The model bias of overproducing frequent low N_d scenarios in MBL clouds in previous E3SM versions remains in E3SMv2 (Varble et al., 2023; Tang et al., 2023; Kang et al., 2024). Satellite LWP in Fig. 1e to 1h varies modestly with a DJF maximum (95.38 g m^{-2}) and a JJA minimum (74.46 g m^{-2}). Spread is greatest in DJF and smallest in JJA. E3SMv2 captures the phase with DJF (91.52 g m^{-2}) and JJA (54.31 g m^{-2}) and matches seasonal means best in DJF and SON, yet the distributions are more positively skewed, indicating a tendency for more frequent low LWP.

Overall, higher N_d and lower LWP in warm seasons and the opposite in cold seasons agree with prior ground-based, satellite, and aircraft evidence (Wu et al., 2020; Varble et al., 2023; Zheng et al., 2024). E3SMv2 successfully captures the broad seasonal variations in both N_d and LWP, but underestimates the amplitude and variability of N_d , especially during the warm seasons, pointing to needed refinements in aerosol cloud interactions and microphysical process representation.

Deleted: layer-mean cloud droplet number concentration (...) and LWP for low-level clouds over the ENA, from satellite (MODIS) retrievals and E3SMv2 model. The annual mean statistics for N_d (LWP)... Annual means are $88.34 \pm 91.68 \text{ cm}^{-3}$ ($81.9 \pm 66.46 \text{ g m}^{-2}$) for MODIS and $66.27 \pm 38.59 \text{ cm}^{-3}$ ($80.31 \pm 68.61 \text{ g m}^{-2}$) for satellite N_d and LWP, and $65.84 \pm 38.59 \text{ cm}^{-3}$ and $77.49 \pm 73.38 \text{ g m}^{-2}$ for E3SMv2.

Deleted: In comparison, the E3SMv2 simulates... reproduces the seasonal variations... trend with N_d peaking in ... JJA (... peak 100.06 cm^{-3})... and reaching ... DJF minimum in DJF (... 49.00 cm^{-3}); however, the magnitude of these seasonal changes is smaller than that in the satellite data. Moreover, the model consistently... but underestimates N_d across all seasons... in every season (Fig. 1a–1d), with particularly significant... the largest low biases during... in MAM 40.5 cm^{-3} and JJA, where the mean differences between the model and observations are 40.5 and 21.83 cm^{-3} , respectively... Satellite observations display broader distributions with higher variability, manifested in larger standard deviations... especially during MAM (96.09 cm^{-3}) and JJA (123.34 cm^{-3}), indicating that the observed N_d encompasses a wider range of cloud conditions, likely influenced by diverse aerosol sources and variable meteorological conditions... In contrast, the E3SMv2 exhibits lower variability, with N_d distributions that are ... generally narrower N_d distributions, with distinct peaks at low concentrations in all seasons except JJA. The model bias of overproducing frequent low N_d scenarios in the ...BL clouds in previous E3SM versions remains in E3SMv2 (Varble et al., 2023; Tang et al., 2023; Kang et al., 2024), possibly related to the model treatment of cloud droplet formation under low turbulence conditions, which is typical in the MBL stratus and stratocumulus (Shan et al., 2024; Wan et al., 2025).

Deleted: Satellite-derived LWP in Fig. 1e–h displays modest seasonal variations, with the highest mean values observed during DJF (95.38 g m^{-2}) and the lowest during JJA (74.46 g m^{-2}). The spread of LWP distribution is the greatest in DJF, smallest in JJA, and intermediate in MAM and SON. E3SMv2 simulates the similar seasonal cycle of LWP, with the highest values in DJF (91.52 g m^{-2}) and lowest values in JJA (54.31 g m^{-2}). The model performs relatively well in reproducing the seasonal mean LWP during DJF and SON, compared to the observations. However, across all seasons, the model may be biased toward producing more frequent low LWP values, suggested by the more positively skewed distributions.

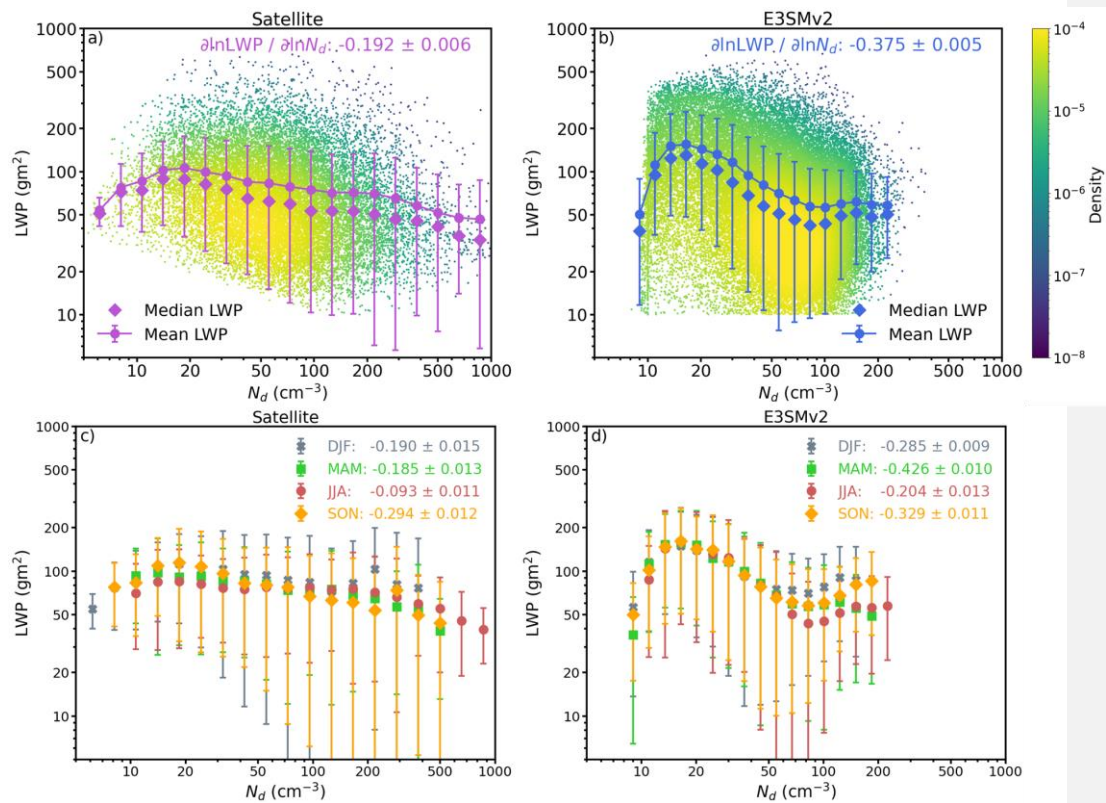


Figure 2. Top Panel: Bulk LWP binned as a function of N_d from a) satellite and b) E3SMv2, the Gaussian kernel density estimate (KDE) is shown as color-shaded scatter points area. The mean (median) LWP values in N_d bins are shown as solid-dotted lines (diamond symbols); Bottom panel: seasonality of the LWP dependences from c) satellite and d) E3SMv2, with colored symbols denote mean LWP in N_d bins. Error bars denote standard deviations of LWP in N_d bins. Slopes are ordinary least squares fits of $\partial \ln(LWP) / \partial \ln(N_d)$, and ‘ $\pm$ ’ values are standard errors of the fitted slopes.

641 The dependence of LWP on N_d for both satellite data and E3SMv2 simulations, is presented in
 642 Figure 2. We quantify the response of LWP to changes in N_d using a LWP adjustment index defined as

643
$$\mathcal{L}_0 = \frac{\partial \ln(LWP)}{\partial \ln(N_d)}.$$

644 We compute $\mathcal{L}_0$ as the slope of an ordinary least squares fit in log-log space between N_d and LWP.
 645 Hence, the $\mathcal{L}_0$ derived from satellite observations and model simulations is -0.192 ± 0.006 and $-0.375 \pm$
 646 0.005 , respectively. The ‘ $\pm$ ’ values reported are the standard errors (SE) of the slope from that fit
 647 (equivalently, 95% confidence level CI = slope ± 1.96 *SE, under standard linear-regression assumptions).
 648 The bulk values are consistent with previous satellite studies over the eastern Atlantic region (Gryspeerd
 649 et al., 2019; Christensen et al., 2023; Zhang et al., 2025). Both datasets show an inverted-V LWP- N_d
 650 relationship. LWP rises with N_d at low N_d then turns negative at higher N_d , with a turning point near 20
 651 cm^{-3} . In the observations (Fig. 2a), increasing N_d suppresses precipitation and allows LWP to accumulate
 652 at low N_d , while at higher N_d the response becomes negative, consistent with enhanced entrainment and
 653 evaporative losses. These results are broadly consistent with prior satellite studies over marine
 654 stratocumulus: an inverted-V LWP- N_d relationship has been reported for N_d ranges of ~ 10 - 300 cm^{-3} in
 655 the southeast Pacific (Goren et al., 2025), globally (Gryspeerd et al., 2019; 2022), and ~ 7 - 400 cm^{-3} for
 656 subtropical stratocumulus (Possner et al., 2020). In contrast, the model simulations appear to generate a
 657 similar, yet exaggerated, shape predominantly through parameterized precipitation suppression
 658 (Mülmenstädt and Feingold et al., 2018; Mülmenstädt et al., 2024b).

659 Global climate models such as E3SMv2 typically lack the resolution to explicitly simulate small-
 660 scale turbulent mixing and entrainment, instead, relying on bulk parameterizations that tend to
 661 overestimate precipitation suppression (Varble et al., 2023; Mülmenstädt et al., 2024a; Y. Zhang et al.,
 662 2024). Noticeably, E3SMv2 (Fig. 2b) captures the relationship between LWP versus N_d qualitatively.
 663 However, the model systematically produces higher LWP at low N_d , and lower LWP at high N_d than
 664 observation, suggesting that E3SMv2 overestimate the sensitivity of LWP to N_d . This discrepancy may
 665 reflect uncertainties in the parameterization of cloud adjustments, particularly those involving
 666 entrainment and aerosol-cloud microphysical interactions in E3SMv2, as discussed further in Section 4.

667 The inverted-V shape in the LWP- N_d relationship persists across seasons in both satellite
 668 observations and E3SMv2 simulations (Figs. 2c and 2d), though its intensity and turning point vary.
 669 Satellite observations (Fig. 2c) generally exhibit a weaker negative slope of LWP with N_d during winter
 670 (DJF) and autumn (SON) than in summer (JJA), with transitional behavior in spring (MAM). In contrast,
 671 E3SMv2 (Fig. 2d) show stronger LWP changes with N_d in every season, consistent with the bulk

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Moved up [1]: et al., 2025).

Deleted: Notably, LWP exhibits a distinct pattern in observations and E3SMv2: it initially increases with N_d and then reverses to a decreasing trend, forming an inverted-V shape with a reversal point near $N_d \sim 20 \text{ cm}^{-3}$. However, while the negative LWP response at high N_d in observations is primarily attributed to entrainment and evaporative processes

Moved up [2]: , with the increased aerosol concentration, cloud droplets become more numerous in smaller sizes. It leads to decrease in efficiency in colliding and coalescing into raindrops and suppress precipitation (Albrecht, 1989). This suppression results in less cloud water being lost through rainfall and, consequently, an increase in LWP. Furthermore, the combined effects of entrainment-sedimentation feedback and precipitation-stabilization may lead to weaker entrainment drying in relatively clean clouds, further enhancing the LWP under lower N_d conditions (Bretherton et al., 2007; Wood, 2012; Possner et al.,

Moved up [3]: Gupta et al., 2021; Zhang et al., 2022; Zheng et al., 2023

Moved up [4]: 2022; Zheng et al.,

Moved up [5]:). This enhancement in evaporation promotes cooling and intensifies localized turbulent mixing, which, in turn, facilitates the entrainment of dry air from above the cloud. The subsequent mixing further accelerates the evaporation of cloud droplets, reducing the overall liquid water content and decreasing LWP.

Deleted: In the satellite observations (Fig. 2a), at lower cloud droplet concentrations

Deleted: 2018). In contrast, at higher N_d levels, the increased abundance of small droplets expands the surface area available for evaporation at the cloud top (Ackerman et al., 2004; ...

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tendency. Despite the overall similarity in seasonal patterns, the discrepancies between the model and observations become more apparent when the data are stratified by season.

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To gauge how satellite retrieval uncertainty affects the LWP versus N_d relationship, we note that satellite N_d studies report normalized root-mean-square differences of about 30 to 50% relative to aircraft data as in Gryspeerd et al. (2022). In log space a multiplicative N_d error is additive in $\ln N_d$, implying an expected slope damping of roughly 10 to 30%. Consistent with this expectation, a Monte Carlo test that multiplied satellite N_d by lognormal noise with coefficient of variation ~ 30 -50% and refit the slope 1000 times reduced the bulk satellite slope from -0.192 to a median of -0.157, an attenuation of about 18%, with a 95 percent sensitivity band of -0.170 to -0.143. However, this does not alter the sign or the comparative result reported here, but it does indicate that observed magnitudes should be interpreted with caution.

3.3 Vertical distribution of aerosol extinction coefficient

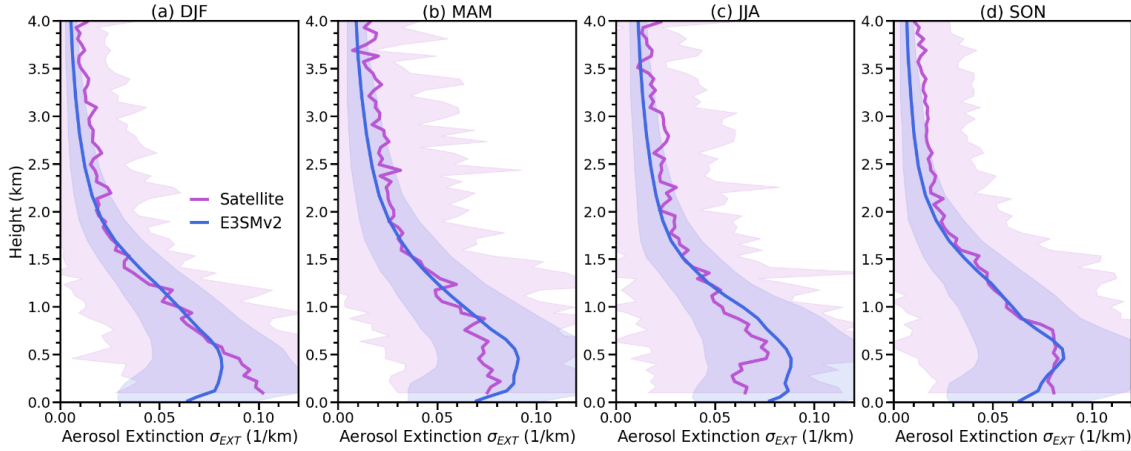


Figure 3. Domain averaged vertical distribution of aerosol extinction coefficients (σ_{EXT}) in the presence of clouds from Satellite (purple) and E3SMv2 (blue) during a) winter, DJF; b) spring, MAM; c) summer, JJA, and d) fall, SON. Shaded area denote the standard deviation of σ_{EXT} for each level.

Figure 3 shows seasonal aerosol extinction profiles over the ENA. Both the satellite record and E3SMv2 feature strongest extinction near the surface below about 1 km, with a rapid drop above roughly 2 km. This steep gradient suggests that the aerosols are more concentrated within the MBL, consistent with surface aerosol sources such as the oxidation of dimethyl sulfide (DMS) and sea spray aerosols (Zheng et al., 2018; Wang et al., 2021; Ghate et al., 2023). Also, the relatively higher relative humidity within the MBL might impacts the optical properties of aerosols (Baynard et al., 2006; Feng et al., 2016).

In winter (DJF) and fall (SON), the model underestimates σ_{EXT} below 1 km compared to satellite observations, whereas in spring (MAM) and summer (JJA) it overestimates near-surface extinction. Recent studies like Logan et al. (2014) and Zheng et al., (2018) demonstrated that the seasonality in the MBL aerosol properties are highly sensitive to local meteorological conditions and long-range transport events. During the cold seasons (DJF and SON), the ENA region experiences high wind speeds in the MBL due to an intensified pressure gradient between the Icelandic Low and the Azores High (Logan et al., 2014; Ghate et al., 2021). The observed high σ_{EXT} in cold seasons reflects coarse-mode contributions from both enhanced sea-salt emissions under strong MBL winds and episodic Saharan dust intrusions that reach the ENA via the synoptic northwestward transport (Logan et al., 2014; Gläser et al., 2015; Rodríguez and López-Darias, 2024). E3SMv2 likely underpredicts this signal due to low sea spray (Burrows et al., 2020) and an underrepresentation of dust vertical extent and transport (Feng et al., 2022), a broader model tendency that appears over the North Atlantic as well (H. Wang et al., 2020; Qin et al., 2024).

In warm months (MAM and JJA), ENA is characterized by enhanced formation of secondary organic aerosols (SOA) and DMS-derived sulfate (Zheng et al., 2018; Sanchez et al., 2018), dominated by fine-mode aerosols, with weaker sea salt under lower winds and reduced dust transport as the Azores High strengthens. (Wang et al., 2021). Conversely, the overestimation of σ_{EXT} by E3SMv2 may be partially attributed to the overproduced sulfate and organic matter at the surface (Hassan et al., 2024; Huang et al., 2024). On the other hand, CALIPSO retrieval limitations under very clean conditions (Painemal et al., 2019) may also contribute to model-observation differences in the free troposphere. Overall, E3SMv2 captures the seasonal evolution of the vertical profile and the mean boundary-layer extinction well enough to support subsequent analysis of simulated cloud responses to aerosol changes.

Deleted: The observed... figure 3 shows seasonal variations in vertically resolved ... aerosol extinction profiles over the ENA are presented in Figure 3.... Both the satellite observations... record and the ... E3SMv2 model display a characteristic decrease in aerosol... feature strongest extinction coefficient with altitude, with maximum values ... near the surface (i.e., ... below approximately... about 1 km) and... with a rapid decline... drop above around... roughly 2 km. This steep gradient suggests that the aerosols are more concentrated within the MBL, consistent with surface aerosol sources such as the oxidation of dimethyl sulfide (DMS) and sea spray aerosols (Zheng et al., 2018; Wang et al., 2021; Ghate et al., 2023). Also, the relatively higher relative humidity within the MBL might also

Deleted: tends to underestimate the aerosol extinction coefficient (... underestimates σ_{EXT})... below 1 km compared to satellite observations, while overestimating the... whereas in spring (MAM) and summer (JJA) it overestimates near-surface extinction during spring and summer.... Recent studies have identified several factors that may explain these seasonal biases. For example,... like Logan et al. (2014) and Zheng et al., (2018) demonstrated that the seasonality in the MBL aerosol properties are highly sensitive to local meteorological conditions and long-range transport events. During the cold seasons (DJF and SON), the ENA region experiences high wind speeds in the MBL due to an intensified pressure gradient between the Icelandic Low and the Azores High (Logan et al., 2014; Ghate et al., 2021). The observed high σ_{EXT} in cold seasons reflects the contribution from ... coarse-mode aerosols, including... contributions from both enhanced sea ... salt emissions under high wind conditions... strong MBL winds and long-range transported... episodic Saharan dust aerosols associated with increased extratropical cyclone activity... intrusions that reach the ENA via the synoptic northwestward transport (Logan et al., 2014; Gläser et al., 2015; Wood et al., 2015). In this context, the underestimation of σ_{EXT} by... Rodríguez and López-Darias, 2024). E3SMv2 may result from underpredicted... likely underpredicts this signal due to low sea spray aerosols and long-range transported dust ... Burrows et al., 2020;... and an underrepresentation of dust vertical extent and transport (Feng et al., 2022), a broader model

Deleted: During spring and summer... n warm months (MAM and JJA), the... ENA is characterized by enhanced formation of secondary organic aerosols (SOA) and DMS-derived sulfate (Zheng et al., 2018; Sanchez et al., 2018), dominated by fine-mode aerosols. Sea salt concentrations, which are usually higher in winter, contribute less during the warm months owing to lower wind speeds. Additionally, as the development of the Azores High in summer inhibits large-scale dust transport to the ENA, thereby reducing contributions from coarse-mode aerosols... with weaker sea salt under lower winds and reduced dust transport as the Azores High strengthens. (Wang et al., 2021). Conversely, the overestimation of σ_{EXT} by E3SMv2 may be partially attributed to the overproduced sulfate and organic matter at the surface (Hassan et al., 2024; Huang et al., 2024). On the other hand, the aerosol retrievals from ... CALIPSO have certain... retrieval limitations at... under very low aerosol... lean conditions (Painemal et al., 2019), which might... may also contribute to the ... model-data... bservation differences in the free troposphere. In general, the ... overall, E3SMv2 well ... aptures the observed ... easonal variations in

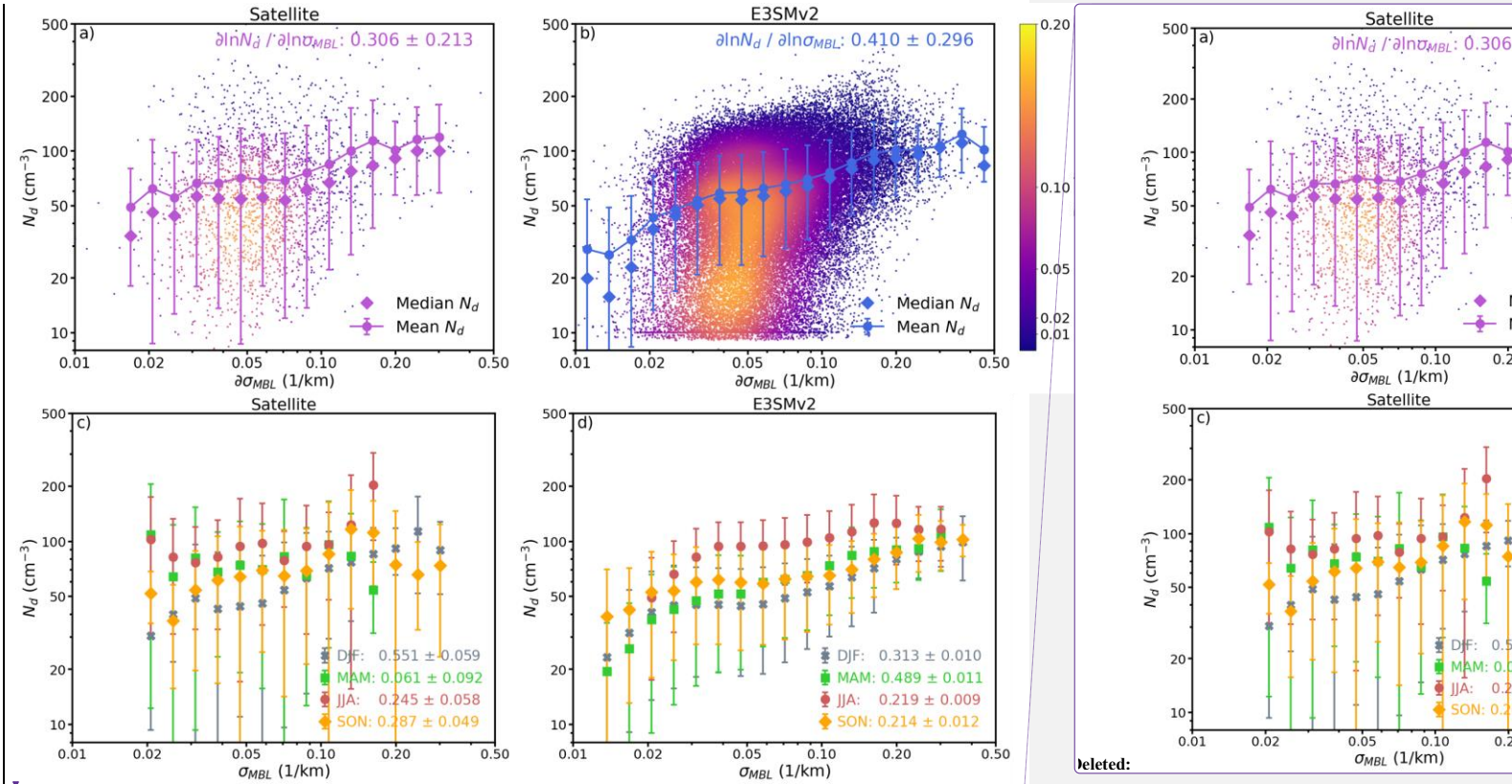


Figure 4. Top Panel: N_d binned as a function of σ_{MBL} from a) satellites and b) E3SMv2. The Gaussian kernel density estimate (KDE) is shown as color-shaded scatter points area. The mean (median) N_d values in σ_{MBL} bins are shown as solid-dotted lines (diamond symbols). Bottom panel: seasonality of the N_d dependences from c) satellite and d) E3SMv2, with colored symbols denote mean N_d in σ_{MBL} bins. Error bars denote standard deviations of N_d in σ_{MBL} bins. Slopes are ordinary least squares fits of $\partial \ln(N_d) / \partial \ln(\sigma_{MBL})$ and '±' values are standard errors of the fitted slopes.

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To quantify the aerosol impact on cloud microphysics, we define an aerosol-cloud interaction (ACI) index as:

$$ACI_N = \frac{\partial \ln(N_d)}{\partial \ln(\sigma_{MBL})},$$

where the σ_{MBL} denotes the mean value of the below-cloud-top σ_{EXT} . This parameter represents the sensitivity of N_d to changes in aerosols within the marine boundary layer (MBL). Aircraft in situ measurements near cloud base provide the most physically robust ACI assessment (Gupta et al., 2021; Zheng et al., 2024). However, it is challenging to do that with satellite data and model outputs, because satellite remote sensing like CALIOP cannot reliably determine cloud-base height, and the model's coarse vertical resolution makes it difficult to collocate the model cloud-base with CALIOP layers. Hence, those factors necessitate the use of the mean aerosol properties within the below-cloud-top MBL in the present study, facilitating a consistent comparative assessment of aerosols between satellite observations and model simulations.

The top row of Figure 4 compares the satellite-derived and E3SMv2-simulated relationships between N_d and σ_{MBL} . Satellites (Fig. 4a) show a moderate N_d sensitivity with considerable scatter ($ACI_N = 0.306 \pm 0.213$). The positive ACI_N reflects the Twomey effect and lies within reported satellite ranges over marine stratocumulus and the eastern Atlantic. For example, McCoy et al. (2017) found a log-log slope of 0.31 between N_d and sulfate mass, Jia et al. 2021 reported 0.14-0.51 for N_d versus AOD over oceans, and recent reviews summarize satellite-based susceptibilities of about 0.1–0.7 depending on sampling and aerosol proxies (Gryspeerd et al., 2023). In comparison, E3SMv2 (Fig. 4b) reproduces the qualitative increase of N_d with σ_{MBL} , but with a noticeably steeper slope, consistent with an overly sensitive microphysical or activation response noted in previous studies (Christensen et al., 2023; Varble et al., 2023). In other words, for the same fractional change in σ_{MBL} , E3SMv2 predicts a larger fractional change in N_d compared to satellite data. This model–observation discrepancy may reflect uncertainties in how E3SMv2 parameterizes aerosol activation, updraft velocities at the cloud base, or boundary-layer processes such as entrainment and mixing (Tang et al., 2024; Wan et al., 2025), as discussed further in Section 4.

In terms of seasonal variations in N_d susceptibility, Satellite derived ACI_N (Fig. 4c) is largest in DJF, near 0.55, and smallest in MAM, near 0.06, with JJA and SON intermediate. E3SMv2 (Fig. 4d) remains positive in all seasons but spans a narrower range, including a much stronger MAM value near 0.49, than observed and a weaker DJF response than satellites, while JJA and SON are closer to observations, yet the model maintains a higher and more uniform sensitivity overall.

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Deleted: can be performed using aerosol information near the cloud base such as from in situ aircraft measurements ... Gupta et al., 2021; Zheng et al., 2022a), limitations in ... (2024). However, it is challenging to do that satellite retrievals of accurate ... data and model outputs, because satellite remote sensing like CALIOP cannot reliably determine cloud ... base height, and the challenge for models to pinpoint the cloud base from ... model's coarse vertical resolution. Moreover, the coarse vertical resolution of the model ... makes it difficult to vertically ... allocate CALIOP aerosol layer and ... the model output ... cloud-base with layers. Hence, those reasons ... actors necessitate the use of the mean aerosol properties within the below-cloud-top MBL in the present study. This approach facilitates a more

Deleted: Satellite observations ... atellites (Fig. 4a) show that N_d generally increases with σ_{MBL} ; however, the ... moderate N_d sensitivity is moderate ... with considerable scatter ($ACI_N = 0.306 \pm 0.213$). This susceptibility falls within the range reported in previous satellite studies (Quaas et al., 2020; Jia et al., 2022), although it is relatively moderate compared to those analyses based on alternative observational platforms such as ground-based observations and in-situ measurements (McComiskey et al., 2009; Zheng et al., 2024). Nonetheless, the ... he positive ACI_N indicates that, on average, higher aerosol concentrations are associated with increased cloud droplet numbers, consistent with the well-known ... effects the Twomey effect.

In contrast, the ... and lies within reported satellite ranges over marine stratocumulus and the eastern Atlantic. For example, McCoy et al. (2017) found a log-log slope of 0.31 between N_d and sulfate mass, Jia et al. 2021 reported 0.14-0.51 for N_d versus AOD over oceans, and recent reviews summarize satellite-based susceptibilities of about 0.1–0.7 depending on sampling and aerosol proxies (Gryspeerd et al., 2023). In comparison, E3SMv2 model ... Fig. 4b) exhibits a stronger dependence of N_d on σ_{MBL} , with a noticeably higher slope. Although the model ... reproduces the qualitative relationship, its ... ncrease of N_d with σ_{MBL} , but with a noticeably steeper slope suggests that the ... consistent with an overly sensitive microphysical or aerosol ... ctivation schemes ... esponse noted in E3SMv2 may be overly sensitive, as also indicated by ...

Deleted: the ACI_N ... atellite derived from satellite data ... ACI_N (Fig. 4c) reveals large differences across seasons: winter (...s largest in DJF) shows a markedly stronger response (~...near 0.55) compared to spring (...and smallest in MAM) at (~...near 0.06). Summer (... with JJA)...and fall (...ON) display ... intermediate ACI_N values. In contrast, the ... E3SMv2 results ... Fig. 4d) also exhibit a clear ... remains positive relationship in every season, ... n all seasons but associated with ... pans a narrower range of ACI_N values. Notably, the simulated springtime (... including a much stronger MAM) ACI_N (~...value near 0.49) is considerably stronger ... than that ... bserved from ... nd a weaker DJF response than satellites, whereas a weaker slope is simulated in winter. In summer (...hile JJA)...and fall (...ON), the slopes ... are closer to satellite-derived values ... bservations, yet E3SMv2 tends to exhibit ... he model maintains a higher more consistent responsiveness of N_d to aerosol increases

1183 Taken together, E3SMv2 tends to produce both stronger LWP responses to N_d and enhanced N_d
 1184 responses to aerosol than satellite-derived relationships. A notable limitation of this seasonal grouping is
 1185 that it fails to disentangle the complexities of aerosol-cloud interactions from characteristic
 1186 meteorological variations. Aggregating data into seasons makes it challenging to unambiguously
 1187 attribute changes in cloud properties to aerosol variations rather than to shifts in large-scale dynamics or
 1188 thermodynamic conditions (Zheng et al., 2022b; Zhang et al., 2023). This limitation necessitates the
 1189 regime-based analysis in Section 4, in which samples are clustered in dominant meteorological regimes,
 1190 and can more effectively isolate microphysical processes from the confounding effects of synoptic-scale
 1191 and seasonal variability (Mülmenstädt et al., 2012; Mechum et al., 2018; Zheng et al., 2025).

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1192 4 Regime-based Analysis of Aerosol, cloud properties, and their interactions

1193 4.1 Distinctive Meteorological Regimes over ENA

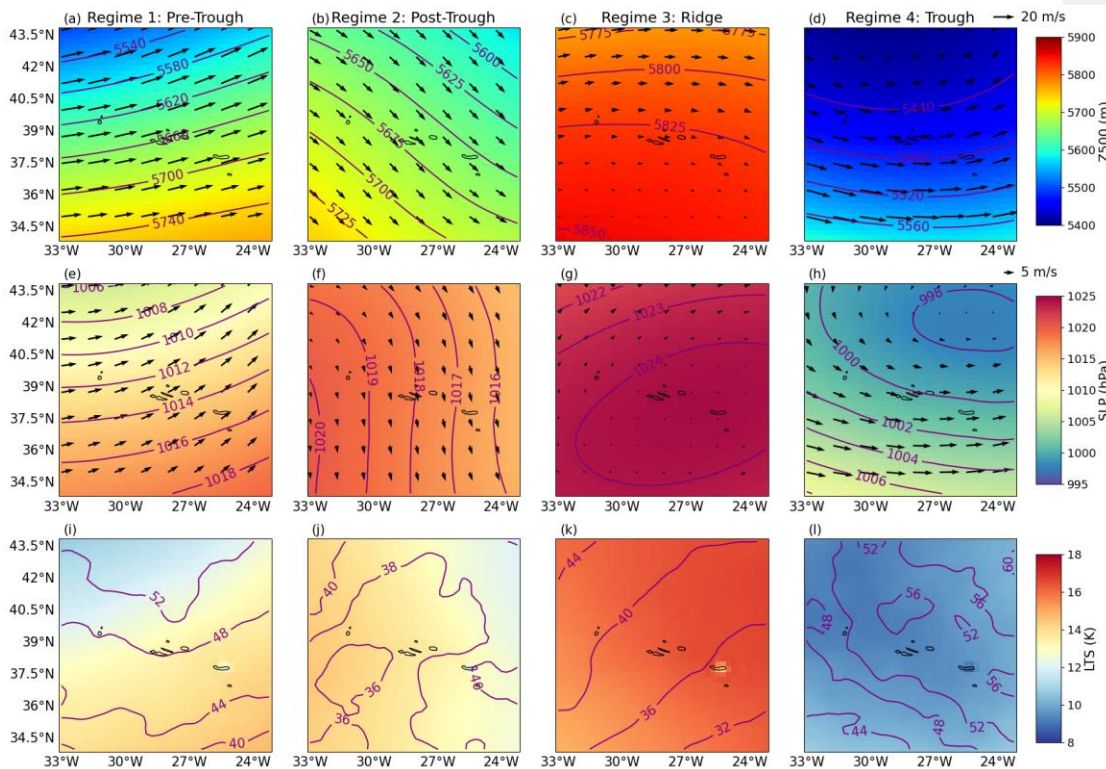


Figure 5. Meteorological composites for each synoptic regime classified by the clustering model: Regime 1 (a, e, i), Regime 2 (b, f, j), Regime 3 (c, g, k), and Regime 4 (d, h, l). The first row shows 500 hPa geopotential height (Z500) shaded and contoured, with 500 hPa wind vectors overlaid. The second row presents sea level pressure (SLP) in both shaded and contoured formats, together with 10 m surface wind vectors. The third row displays lower tropospheric stability (LTS) at 700 hPa as the shaded field and 700 hPa relative humidity (RH700) in contours.

As detailed earlier, the CNN-LSTM-DEC clustering of 3,286 daily ERA5 states results in the identification of four distinct synoptic-scale regimes (Figure 5). Namely, Pre-Trough (regime 1), Post-Trough (regime 2), Ridge (regime 3) and Trough (regime 4). For each regime, composites were computed as the arithmetic mean of the corresponding ERA5 fields across all time steps assigned to that regime.

Regime 1 (Figs. 5a, 5e, and 5i) represents the Pre-Trough phase, characterized by a developing trough approaching the Azores. This regime features strong southwesterly winds at both 500 hPa and near the surface, moderate mid-level moisture, and low lower tropospheric stability (LTS). Such conditions typically precede frontal development and passage and are associated with the early stages of midlatitude cyclone progression, therefore resulting in a wet free troposphere and low LTS (Mechem et al., 2018; Zheng et al., 2025).

Regime 2 denotes the typical Post-Trough condition that follows the passage of a trough, characterized by prevailing northwesterly winds (Fig. 5b) and transitional stability. The SLP field reveals a relatively weak pressure gradient, corresponding to a post-frontal environment in which drier and colder air is advected into the region (Figs. 5f and 5j). Although LTS remains moderate, it is slightly higher than in Regime 1, reflecting the gradual stabilization behind the frontal system. Taken together, Regimes 1 and 2 depict the typical meteorological evolution associated with mid-latitude cyclones traversing the ENA, which occur regularly throughout the year (Table 2), particularly during colder seasons when mid-latitude cyclone activity is more frequent (Wood et al., 2015; Mechem et al., 2018).

Regime 3 corresponds to the Ridge phase, in which a pronounced ridge dominates the region. Both the SLP and 500 hPa geopotential height (Z500) fields constitute a broad anticyclonic pattern, with relatively weak and variable winds from the surface up to 500 hPa. This pattern coincides with the driest free troposphere and the most stable lower troposphere observed among all regimes, which generally favor a more coupled and shallower (MBL) (Carrillo et al., 2015; Zheng et al., 2025). As indicated in Table 2, Regime 3 is the most frequently occurring regime (63.8%) in the region, peaking during the summer months when the center of the Azores High predominantly lies to the southwest of the Azores

Deleted: Large-scale meteorological fields, specifically

Moved up [12]: is based on a combination of silhouette score analysis (i.e., measures of cluster cohesion and separation for different cluster numbers) and the sensitivity of aerosol and cloud property distinctions to the chosen number of clusters. A

Deleted: 500 hPa geopotential height, 10 m horizontal winds, and sea level pressure at 13:00 LT (1 p.m. local time) from the ERA5 reanalysis, are used as inputs to a deep-learning-based...

Deleted: model. The determination of cluster numbers

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Islands. This finding is consistent with previous studies (Mechem et al., 2018; Wang et al., 2022), and the synoptic categorization is similar to that in Painemal et al. (2023) for the Western North Atlantic.

Regime 4 represents a typical Trough phase, characterized by a canonical 500 hPa trough with stronger cyclonic flow at the surface. The lower troposphere exhibits reduced static stability, while the contours of relative humidity at 700 hPa (RH700, Fig. 41) indicate a more humid troposphere. These conditions imply enhanced ascent and moist processes. Note that among the four regimes, Regime 4 is the least frequent (3.4%) and is largely confined to the colder seasons (winter and spring), confirming the findings from previous studies (Wood et al., 2015; Mechem et al., 2018; Wang et al., 2022).

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4.2 Cloud properties under different regimes

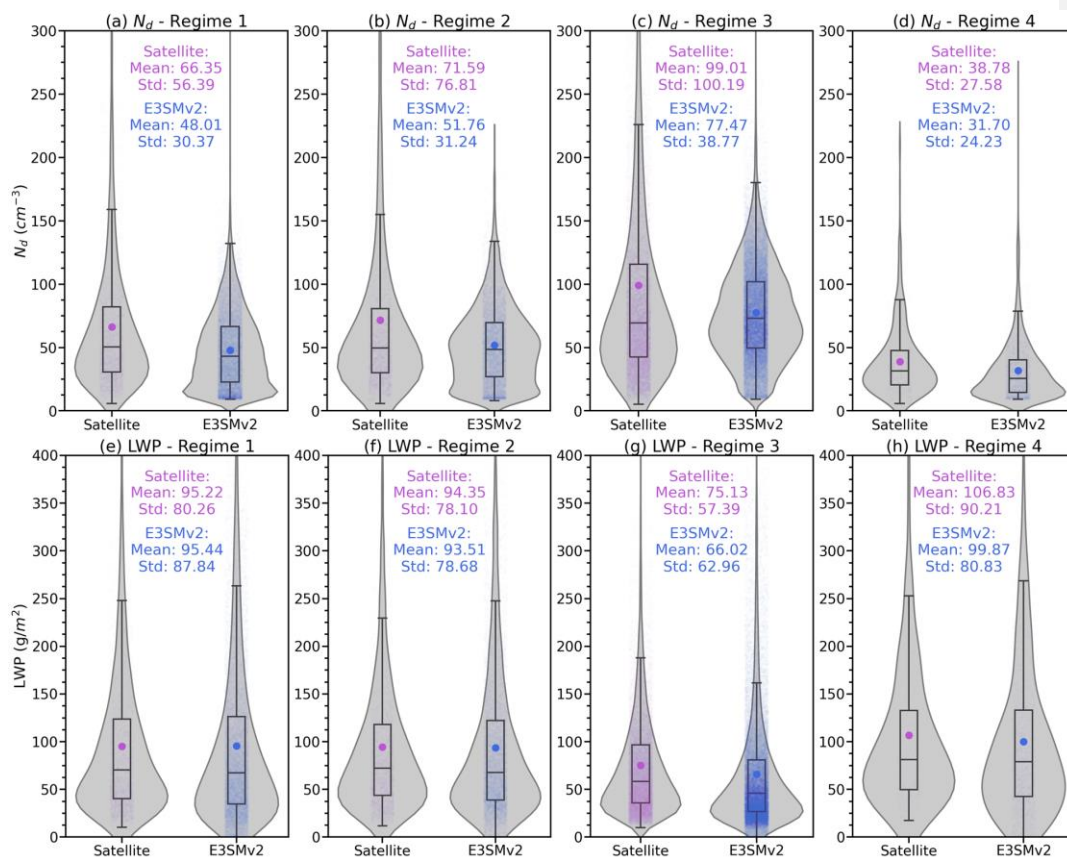


Figure 6. Violin plots of cloud droplet number concentration (N_d , top panels) and cloud liquid water path (LWP, bottom panels) from satellite retrievals (purple) and E3SMv2 simulations (blue), grouped by Regime 1 (a, e), Regime 2 (b, f), Regime 3 (c, g), and Regime 4 (d, h). The mean value is indicated by the color-coded dot. The smoothed shape of each violin shows the Gaussian kernel density estimate (KDE). From top to bottom within each violin, the box plot lines represent the third quartile (Q3, 75th percentile), median (Q2, 50th percentile), and first quartile (Q1, 25th percentile), respectively. The upper whisker extends to $Q3 + 1.5 \times \text{IQR}$ (IQR: interquartile range), and the lower whisker extends to $Q1 - 1.5 \times \text{IQR}$.

In order to provide synergy on the meteorology and cloud for reference in this study, a brief and qualitative summary of meteorology patterns and cloud and precipitation status are listed in Table 1. Furthermore, based on the four distinct meteorological regimes identified through clustering, we stratify cloud properties from both satellite retrievals and E3SMv2 simulations. Both N_d and LWP exhibit systematic regime-dependent behavior and model biases (Fig. 6). And the detailed quantities are listed in Table 3.

Across all regimes, E3SMv2 tends to underestimate N_d while representing LWP more accurately. A rigorous comparison of surface precipitation rates between satellite data and E3SMv2 is limited in this study due to lack of collocated precipitation rate measurements from the satellite. Therefore, we choose to compare the in-cloud fractional occurrences of rain and rain LWP from E3SMv2 with the fractional occurrences of drizzle, light rain, and rain from CloudSat (Table S1). In addition, CTH statistics, stratified by regime from both satellite and E3SMv2 datasets, are presented in Table 2, with composite maps shown in Figures S1 and S2. To quantify the variability of CTH under different regimes, Moran's I indices, a measure of spatial autocorrelation, were computed and then normalized for each regime to account for the differences in spatial resolution between the satellite and E3SMv2 datasets. A normalized Moran's I index of 0 means the least autocorrelated, and 1 means the most autocorrelated. The normalized Moran's I indices for the satellite (E3SMv2) datasets are 0.61 (0.0), 0.64 (0.89), 1.0 (1.0), and 0.0 (0.13) for Regimes 1, 2, 3, and 4, respectively.

Regime 1, characterized by an approaching trough with southwesterly flow, moderate moisture, and relatively weak subsidence, exhibits a mean N_d of 66.35 cm^{-3} in satellite observations, whereas E3SMv2 simulates a lower mean N_d of 48.01 cm^{-3} with reduced variability. The mean LWP in E3SMv2 (95.44 g m^{-2}) is similar to that of the satellite retrievals (95.22 g m^{-2}), with a slightly broader distribution. These findings suggest that while E3SMv2 captures the overall liquid water content in cloud layers under

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pre-frontal conditions, it systematically underestimates N_d . Moreover, the CTH in Regime 1 remains low (Table 3), reflecting a shallower MBL in the transitional environment of the approaching front (Jeong et al., 2022). Precipitation statistics indicate moderate fractions of drizzle and light rain (Table S1), consistent with the notion that although pre-trough instability favors drizzle production, the boundary layer does not fully deepen to support frequent or intense rainfall (Wood, 2005; Wu et al., 2020; Zheng et al., 2022b). The partial uplift and moderate moisture convergence can occasionally enhance cloud thickness, as indicated by a moderately high normalized Moran's I index (0.61), yet the overall shallower structure typically limits heavier precipitation.

In the post-trough environment of Regime 2, the northwesterly flow advects drier and cooler air behind the frontal system. Such conditions are typically associated with increased subsidence and a deeper MBL conducive to the development of stratocumulus clouds (Wu et al., 2020; Jensen et al., 2021; Jeong et al., 2022). Consequently, satellite-derived CTH values (Table 3 and Fig. S1b) are higher than those in Regime 1, indicating a deeper MBL. Furthermore, clouds in Regime 2 are also associated with the highest fractional occurrences of drizzle among the four regimes (Table S1). Drizzle formation and turbulent mixing can lead to heterogeneous cloud structures with less cloud adiabaticity (Wu et al., 2020), as reflected by a normalized Moran's I index of 0.64 and in Figure S1b, in contrast to the more uniform CTH variation in Regime 3. E3SMv2 captures the relative CTH variation and precipitation frequency reasonably well under this regime. As for Regime 1, E3SMv2 still systematically underestimates N_d (51.76 cm^{-3} vs. 71.59 cm^{-3}) while simulating the observed mean LWP well (93.51 vs. 94.35 g m^{-2}).

Regime 3 features with a pronounced ridge, reduced convective activity, and generally lower moisture in the boundary layer. Satellite retrievals show the highest mean N_d (99.01 cm^{-3} , Fig. 6c), which could be attributed to the less active drizzle processes and droplet evaporation in shallower cloud deck within a more stable MBL (Wood et al., 2012; Zheng et al., 2024). In contrast, E3SMv2 simulates a lower mean N_d (77.47 cm^{-3}) and fails to capture the broad observed distribution. Moreover, LWP in this ridge regime is the lowest among all regimes, with E3SMv2 underestimating satellite observations (66.02 vs. 75.13 g m^{-2} ; Fig. 6g). From both satellite and model perspectives, the cloud field is characterized by shallower, more homogeneous decks that span large horizontal areas yet produce only light or sporadic drizzle, as reflected by lower CTH (Figs. S1c and S2c) and the lowest precipitation fractions among the four regimes (Table S1). Overall, the results in Regime 3 align with the signature of shallow stratus and stratocumulus clouds (Rémillard and Tselioudis, 2015; Mechum et al., 2018; Wu et al., 2020; Jensen et al., 2021).

Under a well-developed trough in Regime 4, satellite observations record the lowest N_d (38.78 cm^{-3}) but the highest LWP (106.83 g m^{-2}) among four regimes, reflecting the prevalence of deep

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1316 and warm-rain-active cloud systems formed by strong uplift and abundant moisture. These conditions
1317 yield the highest and most variable CTH among the four regimes (Table 3), along with frequent
1318 precipitation in the form of drizzle or rain. Stronger vertical motion promotes the development of deeper
1319 clouds with higher rainfall efficiency, contributing to spatially heterogeneous precipitating cloud fields
1320 observed in both the satellite data and model simulations (Figs. S1d and S2d; Table S1). Notably,
1321 E3SMv2 simulates the high LWP (99.87 g m^{-2}) and elevated liquid water content as in the observations,
1322 but underestimates variability. Similarly, the model underestimates N_d (31.70 cm^{-3}) relative to satellite
1323 observations as in the other regimes.

1324 Overall, these results demonstrate the clear meteorological impact on both cloud microphysical
1325 (N_d) and macrophysical (LWP) properties. Pre- and post-trough conditions (Regimes 1 and 2) favor
1326 moderate N_d and LWP, while ridge-dominated conditions (Regime 3) promote stable, stratiform-
1327 dominated environments with high mean N_d but relatively low LWP. In contrast, developed troughs
1328 (Regime 4) yield lower N_d yet substantially higher LWP in more vertically developed cloud systems.
1329 Although E3SMv2 captures the LWP mean and distributions across these synoptic regimes, it
1330 systematically underestimates N_d . This discrepancy suggests that the challenges in simulating cloud
1331 droplets are irrespective of the meteorological influences.

Table 1. Summary of meteorological and cloud categories in different regimes.

Regimes	Meteorological Patterns	Cloud Status
R1 Pre-Trough	Approaching trough; strong SW winds at 500 hPa & surface; moderate mid-level moisture; low LTS	Lower CTH; moderate drizzle / light rain
R2 Post-Trough	Post-frontal NW winds; moderate LTS; weak pressure gradient; drier, free troposphere	Higher CTH; high drizzle fraction but low overall precipitation
R3 Ridge	Broad anticyclonic ridge; high-pressure-dominated surface; driest free troposphere; highest LTS	Shallow MBL; Lowest CTH; minimal precipitation
R4 Trough	Canonical trough; strong cyclonic surface flow; lowest LTS; moist free troposphere	Highest & most variable CTH; most frequent drizzle / rain

Table 2. Regime occurrences over ENA at 13:00 LT (1 p.m. local time) from ERA5 inputs

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<u>Count</u> <u>(Fraction %)*</u>	<u>Regime 1</u> <u>Pre-Trough</u>	<u>Regime 2</u> <u>Post-Trough</u>	<u>Regime 3</u> <u>Ridge</u>	<u>Regime 4</u> <u>Trough</u>
<u>Winter (DJF)</u>	<u>269 (33.2%)</u>	<u>166 (20.4%)</u>	<u>313 (38.6%)</u>	<u>63 (7.8%)</u>
<u>Spring (MAM)</u>	<u>193 (23.3%)</u>	<u>169 (20.4%)</u>	<u>424 (51.2%)</u>	<u>42 (5.1%)</u>
<u>Summer (JJA)</u>	<u>26 (3.1%)</u>	<u>22 (2.7%)</u>	<u>780 (94.2%)</u>	<u>0 (0%)</u>
<u>Fall (SON)</u>	<u>111 (13.6%)</u>	<u>122 (14.9%)</u>	<u>581 (70.9%)</u>	<u>5 (0.6%)</u>
<u>Total</u>	<u>599 (18.2%)</u>	<u>479 (14.6%)</u>	<u>2098 (63.8%)</u>	<u>110 (3.4%)</u>

*Percentages in bracket denote the fractional occurrence of specific regime among total sample in each season category (and total sample).

Table 3. Regime-based aerosol and cloud variables from Satellite and E3SMv2

	Regime 1	Regime 2	Regime 3	Regime 4
Satellite				
N_d (cm^{-3})	66.35 ± 56.39	71.59 ± 76.81	99.01 ± 100.19	38.78 ± 27.58
LWP (gm^2)	95.22 ± 80.26	94.35 ± 78.10	75.13 ± 57.39	106.83 ± 90.21
CTH (km)	1.59 ± 0.46	1.76 ± 0.42	1.46 ± 0.50	1.70 ± 0.45
σ_{MBL} ($1/km$)	0.075 ± 0.047	0.070 ± 0.046	0.063 ± 0.039	0.067 ± 0.048
E3SMv2				
N_d (cm^{-3})	48.01 ± 30.37	51.76 ± 31.24	77.47 ± 38.77	31.70 ± 24.23
LWP (gm^2)	95.44 ± 87.84	93.51 ± 78.68	66.02 ± 62.96	99.87 ± 80.83
CTH (km)	1.62 ± 0.84	1.76 ± 0.68	1.41 ± 0.59	1.81 ± 0.88
σ_{MBL} ($1/km$)	0.070 ± 0.038	0.061 ± 0.033	0.071 ± 0.041	0.063 ± 0.032

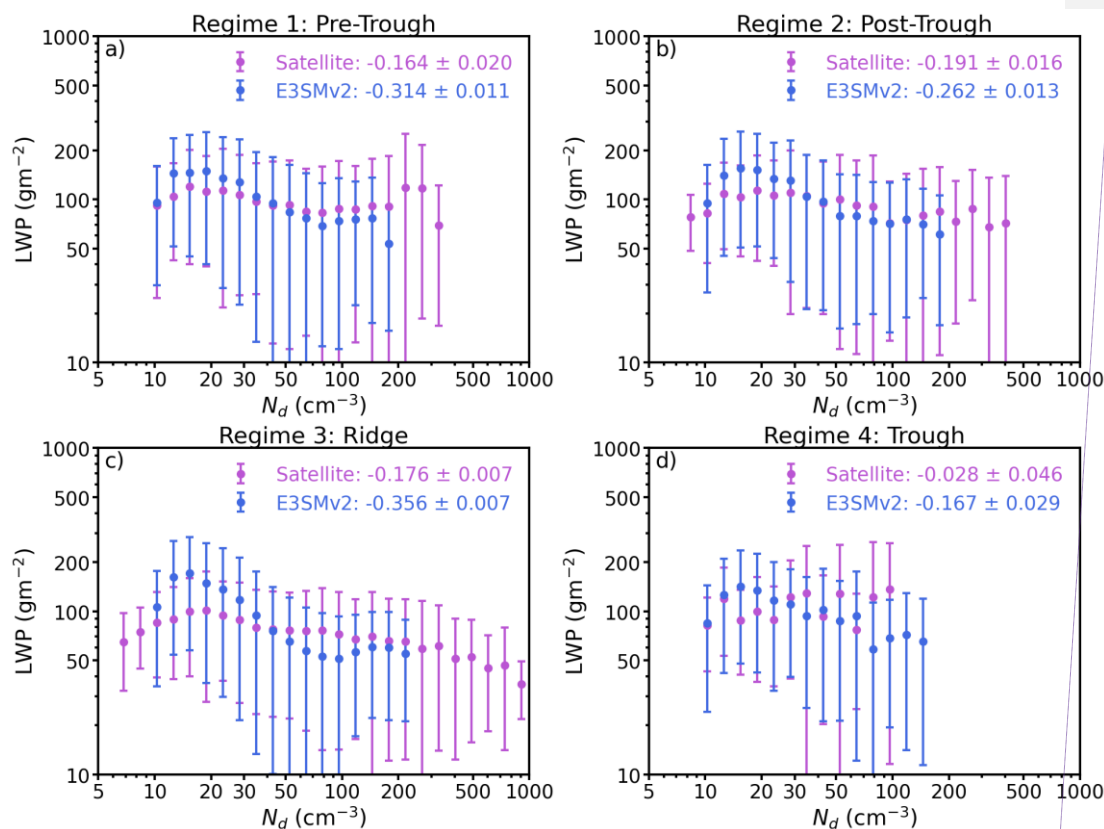


Figure 7. LWP responses on N_d from satellite (purple) and E3SMv2 (blue) for a) Regime 1, b) Regime 2, c) Regime 3, and d) Regime 4. Colored dots denote mean LWP in N_d bins, and whiskers denote standard deviations. The quantitative LWP adjustment index $\mathcal{L}_0 = \partial \ln(LWP) / \partial \ln(N_d)$ is denoted in the legend, and ‘ $\pm$ ’ values are standard errors of the fitted slopes.

The relationship between LWP and N_d across the four synoptic regimes in both satellite retrievals and the E3SMv2 model exhibits the characteristic inverted-V shape (Fig. 7), as shown in the seasonal assessment in Section 3. Furthermore, regime-specific meteorological differences, particularly variations

1347 in stability and moisture transport, strongly influence the shape and peak of the LWP- N_d relationship.
1348 Across regimes, the peak in the satellite data of the inverted LWP- N_d curve occurs at $N_d \approx 15$ -96 cm^{-3}
1349 (particularly, 96.5 cm^{-3} at Regime 4), and at LWP ≈ 101 -136 g m^{-2} ; whereas the E3SMv2 peaks at a
1350 much narrower $N_d \approx 15$ -19 cm^{-3} with higher peak LWP ≈ 142 -171 g m^{-2} . The quantitative LWP
1351 adjustment ($\mathcal{L}_0$) values are listed in Table 4.

1352 Noticeably, satellite retrievals in the Ridge regime (R3) display a more pronounced inverted-V
1353 shape compared to themselves in other regimes, with LWP consistently declining as N_d increases at high
1354 values (Fig. 7c). Under Ridge conditions, strong subsidence limits vertical growth, reduces moisture
1355 convergence, and produces a shallower, more stable cloud layer. Although the mixing rate from
1356 entraining drier free-tropospheric air might not be as intense as in thicker and more turbulent clouds, the
1357 drying effect propagates more efficiently through the thinner cloud layer, exerting a significant impact
1358 (Sanchez et al., 2020; Chun et al., 2023). Furthermore, since the Ridge regime features more high- N_d
1359 conditions, the resulting smaller cloud droplets are more susceptible to entrainment-evaporation, leading
1360 to a more efficient removal of cloud water (Possner et al., 2020). In contrast, the E3SMv2 model
1361 significantly overestimates the maximum LWP, and yields a significantly greater LWP decline with N_d
1362 ($\mathcal{L}_0$ values of -0.356 for model vs. -0.176 for satellite).

1363 Under Regime 1 (Pre-Trough, Fig. 7a), the satellite and model data show a rise in LWP as N_d
1364 increases from very low values, while the characteristic inverted-V shape is partially masked by increased
1365 scatter in the satellite-retrieved LWP at high N_d , likely due to the complexity of drizzle, entrainment, and
1366 mixing processes in the pre-frontal environment. The enhanced drizzle formation, indicated by the
1367 relatively higher drizzle and rain fractions in clouds, can reduce the temperature gradient near cloud-top
1368 via condensation warming, stabilizing the cloud layer and partially offsetting the entrainment cooling
1369 and retaining the LWP. Similar to those in Regime 3, the model also exhibits a nearly double $\mathcal{L}_0$ (-0.314)
1370 compared to satellite (-0.164). One possible explanation is that the model microphysics suppresses
1371 drizzle too aggressively when N_d increases occur at lower values (Varble et al., 2023; Mülmenstädt et al.,
1372 2024b), thereby promoting the accumulation of cloud liquid water prior to the onset of entrainment-
1373 drying or precipitation. Furthermore, the MG2 microphysics scheme may trigger the entrainment-
1374 evaporation feedback too rapidly in the model, resulting in an LWP adjustment timescale much shorter
1375 than observed (Zhou et al., 2025). As a result, the model depletes cloud liquid water too quickly (Xie et
1376 al., 2018; Rasch et al., 2019). These combined model uncertainties lead to an excessively steeper $\mathcal{L}_0$
1377 compared to satellite observations.

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1379 In the Post-Trough regime (Regime 2; Fig. 7b), the residual dynamic forcing from the trough
1380 coupled with the onset of entrainment-induced evaporation, produces an intermediate LWP- N_d
1381 sensitivity, particularly under the high N_d condition, between the Pre-Trough and Ridge regimes. On the
1382 one hand, in the Post-Trough environment, the cloud field is more variable and relatively thicker, which
1383 is found to be favorable for stronger entrainment rates (Wood, 2007; Wu et al., 2020; Chun et al., 2023),
1384 hence depleting LWP. On the other hand, this effect might be closely intermingled with patches of high
1385 LWP sustained by the precipitation-stabilization effect (Possner et al., 2020; Wu et al., 2020). These
1386 competing effects result in an averaged LWP decline with increasing N_d that is stronger than in the Pre-
1387 Trough state but remains less pronounced than the sharp decrease observed in the Ridge regime.
1388 Compared to the satellite observations, E3SMv2 underestimates LWP at high N_d , coupled with the
1389 slightly steeper LWP- N_d slope. The model bias likely results from parameterized entrainment and in-
1390 cloud mixing (Y. Zhang et al., 2023), which may intensify cloud-top evaporation cooling and promote
1391 LWP loss; in contrast, real clouds exhibit more heterogeneous mixing that better preserves liquid water
1392 at high N_d .

1393 In Regime 4 (Fig. 7d), characterized by a well-developed trough with enhanced ascent and high
1394 moisture availability, clouds deepen, and LWP peaks at relatively high N_d , as observed by satellites. The
1395 muted LWP decrease seen in satellite data may be largely a consequence of combined factors. Since the
1396 satellite N_d is derived assuming an adiabatic vertical profile and a constant N_d throughout the cloud, the
1397 expected increase in subadiabaticity for Regime 4, as suggested by the enhanced precipitation, might
1398 induce retrieval bias that dampens the apparent sensitivity of LWP to N_d (Grosvenor et al., 2018). At the
1399 same time, retrievals tend to average over heterogeneous cloud fields in the Trough regime, where cloud
1400 properties vary on small scales, further smoothing out the true microphysical sensitivity (Gryspeerdt et
1401 al., 2022). Excluding the retrieval-induced bias, strong updrafts in this regime can promote moisture
1402 convergence, which in turn maintain or even enhance LWP despite higher N_d (Goren et al., 2018; Zhang
1403 et al., 2022; Painemal et al., 2023). In some cases, heavy precipitation may also drive locally
1404 precipitation-generated cold pooling that enhances the cloud base updraft that helps maintain LWP
1405 following precipitation. Such muted LWP response to N_d , has been reported for the stratocumulus clouds
1406 over the southeast Pacific, particularly in thickening stratocumulus clouds with a higher likelihood of
1407 producing more intense precipitation (Smalley et al., 2024), suggesting the precipitation-suppressing
1408 overwhelms the entrainment-drying effect on LWP. In contrast, E3SMv2 maintains an evident inverted-
1409 V shape of LWP- N_d curve as in other regimes, albeit with a smaller $\mathcal{L}_0$ (-0.167), resulting from the
1410 explicitly parameterized microphysical processes.

Overall, the model simulations exhibit an excessively sharp rise-and-fall pattern in LWP, producing an exaggerated inverted-V in the LWP– N_d relationship, compared to the more subtle shapes in satellite results. This suggests that microphysical feedback to cloud water may be triggered too early and too rapidly in the model. Such behavior may originate from the MG2 microphysics scheme’s nonlinear autoconversion rate (Gettelman and Morrison, 2015), which acts to suppress drizzle too aggressively at low N_d , thereby retaining excess LWP, but truncates liquid water accumulation as N_d increases (Wang et al., 2023; Ovchinnikov et al., 2024). These limitations hinder E3SMv2’s ability to capture observed cloud feedback, particularly in regimes with shallower clouds (e.g., Pre-Trough and Ridge). Addressing these issues may require recalibrating autoconversion rates using observational constraints (e.g., ARM data) and improving scale-aware entrainment schemes that better differentiate mixing regimes.

4.4 N_d susceptibility to aerosols under different regimes

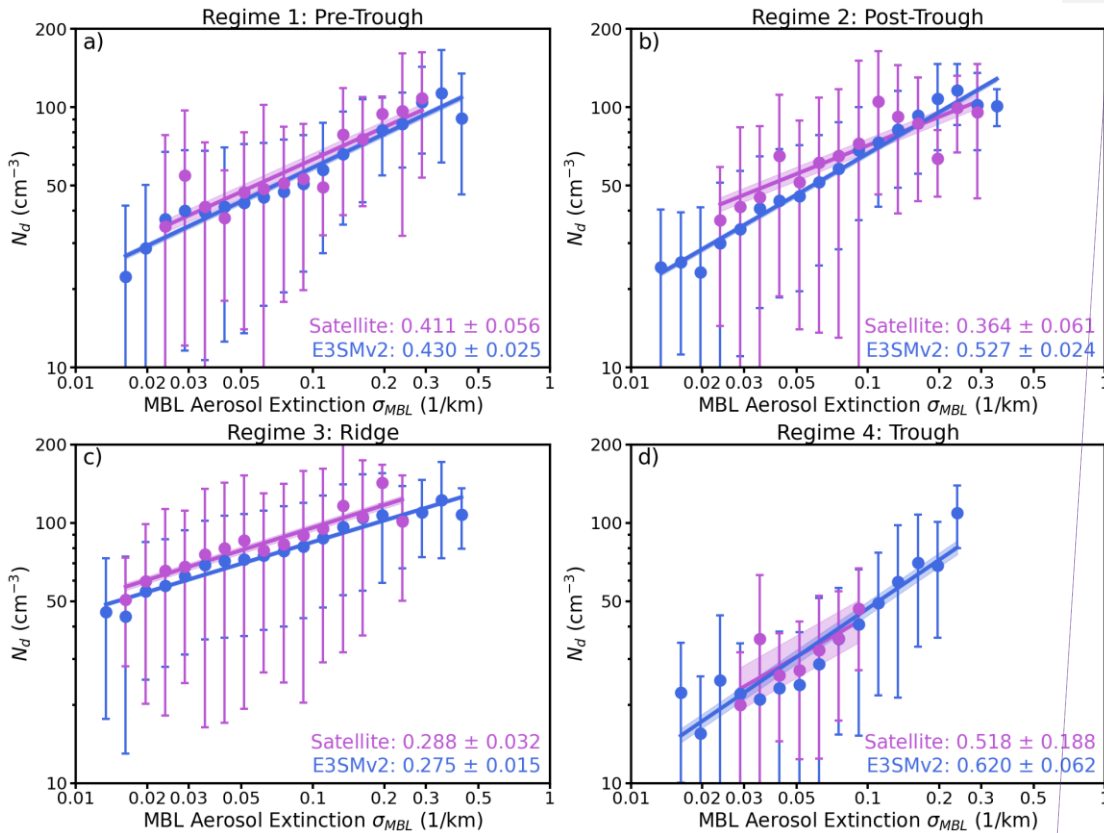


Figure 8. Cloud droplet number concentrations (N_d) dependence on the mean MBL aerosol extinction coefficient (σ_{MBL}) from satellite retrievals (purple) and E3SMv2 simulation (blue), under a) Regime 1; b) Regime 2; c) Regime 3 and d) Regime 4. The solid line indicates the regression line, and the N_d susceptibility $ACI_N = \partial \ln(N_d) / \ln(\partial \sigma_{MBL})$ is denoted in the legend, and ‘ $\pm$ ’ values are standard errors of the fitted slopes.

Figure 8 illustrates how N_d responds to changes in MBL aerosol extinction (σ_{MBL}) across each meteorological regime, as quantified by the aerosol–cloud interaction index (ACI_N). Under all regimes, both observations and E3SMv2 simulations show that N_d generally increases with increasing σ_{MBL} , reflecting the typical ACI in which cloud droplet concentration rises with increased aerosol loading. And it is worth noting that the model is able to simulate the quantitative relationship between N_d and σ_{MBL} , with exception of Regime 2. The ACI_N values are listed in Table 4. Since each regime is characterized by distinct large-scale meteorological conditions, these environmental factors influence the ACI remarkably, as the aerosol activation is highly influenced by updraft strength, moisture availability, and in-cloud supersaturation (Chen et al., 2011; Kirschler et al., 2022; Zheng et al., 2024).

Both satellite retrievals and E3SMv2 yield the lowest N_d susceptibility under the Ridge scenario (Regime 3, Fig. 8c), consistent with the stabilizing effect of subsidence that suppresses updraft variability and limits cloud depth. Furthermore, solar heating on cloud top, especially prominent in the local afternoon, can offset the longwave radiative cooling, thereby stabilize the cloud layer and reduce in-cloud supersaturation (Wood, 2012; Zheng et al., 2018), hence dampening the N_d susceptibility to aerosols. Such mechanism can exert a larger influence on Regime 3, as it is dominated by the warm season (summer and fall) occurrences. The good agreement between the model and satellite observations under these steady conditions suggests that, when cloud-top mixing and convective vigor are limited, E3SMv2 aerosol activation and microphysics perform reasonably well.

Conversely, under trough conditions (Regime 4), retrievals and E3SMv2 produce the highest N_d susceptibility among all regimes (Fig. 8d). In these conditions, stronger updraft and abundant moisture produce deeper MBL clouds, and effectively increase the in-cloud supersaturation (Gong et al., 2023). Therefore, the environmental conditions and the relatively less N_d provide aerosols a greater opportunity to modulate droplet numbers (Hudson and Noble, 2014; Zheng et al., 2024). The E3SMv2 exhibits a steeper slope than that observed, suggesting that the model may overestimate the efficiency with which additional aerosol activates into new droplets, though the discrepancy is less pronounced than those in Regime 2.

1449 Interestingly, the largest overestimation of ACI_N by E3SMv2 occurs under Post-Trough
1450 conditions (Regime 2, Fig. 8b). In this regime, the model exhibits a much higher ACI_N than observations,
1451 suggesting that the model either overestimates aerosol activation or underrepresents processes that limit
1452 droplet concentration, such as drizzle formation and entrainment mixing. In thicker MBL clouds, the
1453 greater vertical extent may allow for enhanced droplet recirculation and collision-coalescence, resulting
1454 in a reduction of N_d that dampens aerosol effects (O et al., 2018; Zheng et al., 2024). Under Pre-Trough
1455 conditions (Regime 1, Fig. 8a), the model slightly overestimates N_d susceptibility yet agrees better with
1456 observations compared to Post-Trough. Previous studies have shown that E3SMv1 exhibits greater
1457 sensitivity of N_d to aerosols than do ground-based and satellite observations (Christensen et al., 2023;
1458 Varble et al., 2023), and this issue appears to persist in E3SMv2 over the ENA, consistent with Huang et
1459 al. (2024). It is possible that the model does not accurately represent the postfrontal boundary layer,
1460 possibly due to the unresolved subgrid turbulence (Ma et al., 2022), which may lead to an exaggerated
1461 N_d response to aerosol extinction.

1462 Note that under low MBL aerosol (σ_{MBL}) conditions, the model yields more occasions of low N_d
1463 compared to the satellite, though it can be due to the limited sample sizes in the satellite, yet the N_d
1464 increases with σ_{MBL} are also more subtle, especially in the Post-Trough and Ridge regimes. Previous
1465 studies suggest that when environmental conditions limit activation and updraft, the resulting droplet
1466 numbers are systematically low (Tang et al., 2023; Varble et al., 2023). In other words, the model
1467 parameterizations lead to under-activation of aerosols in low-aerosol or weak-turbulence regimes. In
1468 contrast, sensitivity experiments by Wan et al. (2025) show that while stronger updrafts can boost N_d ,
1469 doing so would undesirably increase the effective radiative forcing. Therefore, the model aerosol
1470 activation scheme might be oversensitive to the environmental factors, as shown in the present study. It
1471 is also noteworthy that E3SMv2 simulates a greater extent of σ_{MBL} than is observed by satellites under
1472 all scenarios, particularly in Regime 4, where precipitation is more prevalent. This discrepancy may arise
1473 from insufficient drizzle scavenging at moderate to high aerosol loads (Shan et al., 2024), whereby the
1474 model fails to effectively remove aerosols, rendering it overly sensitive to incremental changes in σ_{MBL} .

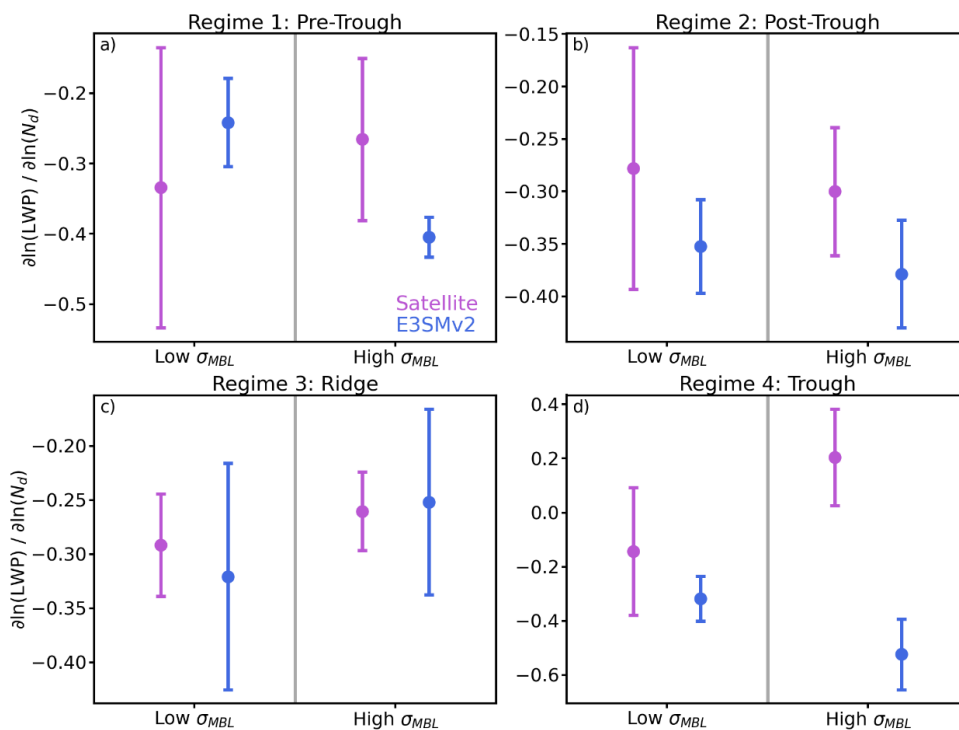


Figure 9. LWP adjustment due to N_d under low and high σ_{MBL} categories, separated by the median σ_{MBL} values (gray line) from the aggregate satellite (purple) and E3SMv2 (blue) dataset. For a) Regime 1; b) Regime 2; c) Regime 3; d) Regime 4. Error bars denote standard errors of fitted slopes in each category.

1476 In order to further illustrate the impact of aerosols on the behavior of the LWP- N_d relationship,
 1477 both satellite and E3SMv2 data are grouped into lower and higher half σ_{MBL} categories, defined by the
 1478 pooled median of the combined satellite and E3SM samples (0.594), and this single threshold is applied
 1479 to all regimes and both datasets to ensures an identical conditioning (Fig. 9).

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1480 In the Pre-Trough regime (Fig. 9a), clouds exhibit low tops and moderate precipitation, featuring
 1481 a regime where precipitation suppression competes with entrainment drying. Satellite retrievals show a
 1482 weaker negative LWP- N_d sensitivity under high aerosol loading (i.e., LWP declines less steeply with
 1483 N_d), whereas E3SMv2 simulates a steeper negative slope. The satellites may observe a weaker decline in

LWP with increasing N_d because moderate precipitation in this regime allows sub-cloud drizzle evaporation to moisten the boundary layer, which weakens the in-cloud humidity gradient and reduces the entrainment efficiency, thereby buffering LWP losses (Wang et al., 2010; Chen et al., 2011; Jia et al., 2022). In the Ridge regime (Regime 3; Fig. 9c), characterized by shallow clouds and minimal precipitation, both satellite retrievals and E3SMv2 simulations yield a relatively weak (less negative) LWP- N_d sensitivity under high aerosol conditions. At higher aerosol loadings, increased N_d suppresses precipitation more effectively in these shallow clouds, stabilizing LWP by limiting drizzle loss. This mechanistic alignment between observations and E3SMv2 may explain their convergence toward a weaker negative slope (Quaas et al., 2020; Jia et al., 2022).

Post-Trough clouds (Regime 2; Fig. 9b), with greater cloud-top heights and moderate precipitation, exhibit a steeper negative LWP- N_d sensitivity in both observations and E3SMv2. The greater depth of Post-Trough clouds amplifies the vertical moisture gradient between the cloud layer and the overlying dry free troposphere. In this scenario, the primary mechanism reducing LWP is enhanced entrainment-driven evaporation. Once droplets are smaller (high N_d), they evaporate faster at cloud top, enhancing evaporative cooling. This strengthens entrainment through buoyancy reversal, creating positive feedback that accelerates LWP loss (Gryspeerd et al., 2019). Although precipitation suppression can still play a role, the net result in a deeper cloud is often dominated by entrainment drying rather than by the retention of liquid water. Consequently, as aerosol loading increases, the slope of LWP- N_d becomes more negative, aligning with findings that deeper clouds, with stronger inversions, exhibit steeper negative slopes in LWP- N_d (Zhang and Feingold, 2023).

The Trough regime (Fig. 9d), marked by deep, precipitating clouds and unstable conditions, highlights a key model-observation discrepancy: satellites detect a weak or even positive LWP- N_d relationship at high aerosol loadings, while E3SMv2 simulates a steeply negative slope. It could be possible that, under the moist and unstable environment, the increasing N_d provides more surface areas for water vapor condensation and hence offsets the entrainment drying loss (Gryspeerd et al., 2019). Also, under this regime with relatively low N_d , the increasing evaporation is favorable for more latent heat release, allowing the cloud to be invigorated and expand in vertical extent, hence increasing the LWP (Altaratz et al., 2014). Moreover, satellite retrievals in this regime may be also biased by vertical cloud inhomogeneity and drizzle contamination, which could artificially inflate LWP estimates in high- N_d conditions (Zhang et al., 2022). Meanwhile, E3SMv2 relies on parameterizations for turbulence and entrainment that are calibrated for shallow stratocumulus and may not fully capture the intermittency of entrainment in thicker, more precipitating cloud regimes (Gettelman and Morrison, 2015; Wang et al., 2023; Tang et al., 2023). This may cause the model to amplify entrainment drying relative to what might

1520 be observed, thereby producing a more strongly negative LWP– N_d slope than indicated by satellites.
1521 Ultimately, these contrasting signals reflect both retrieval limitations in complex cloud systems and the
1522 model’s sensitivity to microphysical closure assumptions (Christensen et al., 2023).

1523 Such model-satellite discrepancies are further confirmed in the bulk indirect susceptibility, which
1524 quantifies the integrated response of LWP to changes in marine boundary layer aerosol extinction. As
1525 shown in Table 4, satellite-derived susceptibilities range from -0.070 to -0.015 , while E3SMv2
1526 simulations consistently yield larger negative values (from -0.138 to -0.098), especially for Regime 2
1527 and 3, which feature more stratiform-like clouds. That is, for a given increase in aerosol loading, E3SMv2
1528 predicts a stronger LWP reduction than satellite observations. This systematic overestimation by
1529 E3SMv2 indicates that the model may be overly sensitive to aerosol perturbations, translating into an
1530 exaggerated indirect effect on LWP. Potential causes for this discrepancy include overly aggressive
1531 drizzle suppression in the MG2 microphysics scheme, which may retain excess LWP at low N_d but
1532 prematurely truncate water accumulation as N_d increases. In addition, limitations in representing subgrid-
1533 scale turbulent mixing and entrainment could further contribute to the observed biases. While E3SMv2
1534 captures the qualitative trends of aerosol–cloud interactions, the quantitative discrepancies highlight the
1535 need for further refinement in aerosol activation and cloud microphysical parameterizations, as well as
1536 improved process-level representations of drizzle processes and entrainment mixing. Addressing these
1537 issues is essential for enhancing the model ability to simulate the indirect effects of aerosols on cloud
1538 properties in the ENA region.

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Table 4. Indirect susceptibility of LWP to Aerosol for Satellite and E3SMv2

	Regime 1	Regime 2	Regime 3	Regime 4
$\frac{\partial \ln(LWP)}{\partial \ln(N_d)}$				
Satellite [*]	-0.164 ± 0.019	-0.191 ± 0.016	-0.176 ± 0.007	-0.028 ± 0.046
E3SMv2 [*]	-0.314 ± 0.011	-0.262 ± 0.013	-0.356 ± 0.007	-0.167 ± 0.029
$\frac{\partial \ln(N_d)}{\partial \ln(\sigma_{MBL})}$				
Satellite [*]	0.411 ± 0.056	0.364 ± 0.061	0.288 ± 0.032	0.518 ± 0.188
E3SMv2 [*]	0.430 ± 0.025	0.527 ± 0.024	0.275 ± 0.015	0.616 ± 0.062
$\frac{\partial \ln(LWP)}{\partial \ln(N_d)} * \frac{\partial \ln(N_d)}{\partial \ln(\sigma_{MBL})}$				
Satellite [†]	-0.067 ± 0.012	-0.070 ± 0.013	-0.051 ± 0.006	-0.015 ± 0.024
E3SMv2 [†]	-0.135 ± 0.009	-0.138 ± 0.009	-0.098 ± 0.006	-0.104 ± 0.021

^{*}Values are slopes from ordinary least squares fits in log–log space. The ‘±’ entries are standard errors.

[†]The errors for the product is obtained by propagating the standard errors of the component $(\partial \ln LWP / \partial \ln N_d)$ and $\partial \ln N_d / \partial \ln \sigma_{MBL}$ slopes $((N_d) / \partial \ln(\sigma_{MBL}))$ multiplicative propagation).

5. Summary and Discussions

This study investigated aerosol-cloud interaction processes over the ENA by assessing satellite retrievals and simulations from E3SMv2. Using newly developed CALIPSO-derived vertical-resolved aerosol extinction and the collocated MODIS cloud properties, along with model output from a 1° nudged E3SMv2 simulation over a ~10°×10° domain from 2006 to 2014. Leveraging a novel regime-based evaluation framework that places E3SMv2 and satellite observations side-by-side within the clustered synoptic regimes, the variations of marine low-cloud and aerosol properties, and the cloud responses to aerosols are examined.

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Our analysis reveals distinct seasonal variations in cloud and aerosol properties, with ~~satellites~~ and ~~E3SMv2 exhibit~~ higher N_d ~~with~~ lower LWP in warm seasons and lower N_d ~~with~~ higher LWP in cold seasons. E3SMv2 ~~consistently~~ underestimates N_d ~~while producing~~ more comparable LWP ~~in line with~~ ~~prior work~~. In general, satellite retrievals and E3SMv2 simulations capture the qualitative trends where higher N_d under increased aerosol loading and a characteristic inverted-V relationship between LWP and N_d . However, while the LWP response to N_d in the satellite dataset is primarily attributed to precipitation suppression and entrainment and evaporative processes, the model simulations ~~generate~~ a similar but more dramatic shape due to deficiencies in representing these processes within the model parameterizations. Furthermore, it is possible that satellite retrieval biases and sampling strategies may contribute to the observed inverted-V behavior in the LWP- N_d relationship (Grosvenor et al., 2018; Arola et al., 2022; Gryspeerdt et al., 2022), ~~though~~ we addressed retrieval uncertainties to some extent by applying several data screenings documented in Painemal et al. (2020). Another possibility is that the inverted-V shape ~~may~~ reflects natural spatial variability that leads to both increases and decreases of N_d with LWP (Goren et al., 2025).

~~To better isolate meteorological controls~~, we employ a deep-learning-based clustering method to partition the ENA meteorology into four distinct synoptic regimes. ~~Namely, Pre-Trough (regime 1), Post-Trough (regime 2), Ridge (regime 3) and Trough (regime 4)~~. This clustering approach ~~captures~~ spatial and temporal variability ~~more effectively than~~ traditional methods ~~and reveals~~ that ~~meteorology strongly modulates~~ aerosol-cloud interactions. The ~~inverted-V LWP- N_d relationship persists~~ across regimes, with regime-dependent ~~differences~~ between ~~satellites~~ and E3SMv2.

Regime 3 (Ridge) shows the strongest simulated inverted-V response among all four regimes, where LWP ~~decreases~~ at high N_d , driven by the stronger contribution of the entrainment-drying effect in a thinner, more stable cloud layer. In the pre-trough regime, the LWP decline with high N_d is partially masked by the interplay between precipitation-stabilization and entrainment-mixing processes. In the Post-Trough regime, deeper clouds with intermediate precipitation display a more negative LWP- N_d sensitivity due to enhanced entrainment-drying effect, a process the model appears to overestimate compared to observations. Conversely, the Trough regime, characterized by deep, precipitating clouds, features complex behavior where satellite retrievals yield a muted LWP response, likely because of precipitation-suppressing overwhelming the entrainment-drying effect on LWP. The model overpredicts the LWP responses across all regimes with different magnitudes. ~~Both~~ datasets show ~~increasing~~ LWP with N_d at low N_d ~~but~~ E3SMv2 ~~peaks at a lower N_d~~ , suggesting ~~overly aggressive drizzle suppression~~ and ~~insufficient representation of~~ subgrid moisture variability. Moreover, the E3SMv2 simulates steeper

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declines of LWP with N_d at higher values, indicating that the model parameterization induces more rapid entrainment-drying effects contributing to excessive LWP loss.

Both satellites and E3SMv2 show N_d increasing with MBL aerosol extinction in every regime. However, model N_d susceptibility varies more with meteorology than the satellite record. In the regimes with relatively shallow clouds, both data and model exhibit the lower N_d susceptibility to aerosol. In contrast, E3SMv2 shows a markedly steeper N_d response than observed in regimes with deeper and more precipitating clouds, likely due to an overestimation of aerosol emission and activation or underrepresentation of limiting processes such as insufficient drizzle scavenging. Such discrepancies suggest that the parameterizations in model aerosol activation schemes might be overly sensitive to environmentally controlled factors, which leads to a larger range of ACI_N .

The regime-wise LWP responses on N_d are analyzed under low and high MBL aerosol extinction conditions. In shallower and less precipitating cloud conditions, such as the Ridge regime, both satellite retrievals and E3SMv2 simulations converge to a weaker (less negative) sensitivity under high aerosol loading. By contrast, in more precipitating and vertically extended regimes, E3SMv2 exhibits significantly stronger LWP depletions on N_d with increasing aerosols. The E3SMv2 operates at relatively coarse horizontal and vertical resolutions, and the parameterized microphysics. Previous sensitivity studies indicate that changes in low clouds from E3SMv2 are noticeably controlled by CLUBB, and followed by MG2, tunings (Zhang et al., 2023). And the MG2 scheme, tends to overemphasize the entrainment-driven drying and droplet evaporation as it was calibrated for shallow stratus and stratocumulus conditions (Tang et al., 2024). As a result, the transition from shallow to deep cloud regimes, where natural processes evolve continuously, may not be adequately captured, leading to an exaggerated drying signal in the model. While independent analysis of the MG2 scheme show bias in warm rain processes, as it realizes the negative LWP pathway too rapidly and strongly (Zhou et al., 2025), implicating the turbulence-microphysics coupling as a persistent bias source that aligns with our regime-specific over-depletion of LWP at high N_d . Regarding model resolution, vertically resolved physics and concurrent horizontal and vertical refinement improve the representation of entrainment mixing processes and reduce stubborn stratocumulus biases (Lee et al., 2022; Bogenschütz et al., 2023). Hence, we cautiously attribute the potential E3SMv2 discrepancies versus satellite results to those simulated processes in the model, while acknowledging that they can be also the combined effects from multiple feedback and interplay between the model schemes.

It is noteworthy that previous ACI studies in a synoptic context have been largely cyclone-centric (e.g., McCoy et al., 2020; Lee et al., 2025). Our regime-stratified results are consistent with that literature: in cyclone-associated conditions (Pre-Trough, Trough) we see LWP increases or smaller decreases with

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1677 higher Nd, whereas in the anticyclonic conditions (Ridge) LWP decreases markedly with higher Nd, as
 1678 expected in stable, dry high-pressure environments. Our clustering approach extends the synoptic pattern
 1679 classification by providing a flexible, data-driven identification that captures the same physical contrasts
 1680 as cyclone masks while explicitly considering the other two regimes (Ridge and Post-Trough). Therefore,
 1681 our approach might be more general, and remaining applicable beyond the regions dominated by
 1682 midlatitude storm tracks. Model behavior also parallels prior findings: E3SMv2's overly steep LWP
 1683 reductions in Ridge conditions mirror the overestimation of LWP sensitivity outside cyclones reported
 1684 by McCoy et al. In short, our aim is to develop a data-driven way to untangle meteorology from cloud
 1685 responses without pre-specifying synoptic systems, and the learned regimes would naturally recover the
 1686 traditional cyclone phases (pre-, post-trough, trough). And we also view this as transferable to other
 1687 regions of the globe, including marine stratocumulus regions with weaker cyclone influence (e.g., the
 1688 southeastern Atlantic).

1689 In summary, our findings report the satellite-observed and model-simulated range of aerosol-
 1690 cloud interaction indices from both seasonal and regime-based perspectives over the ENA. Moreover,
 1691 the regime-based analysis demonstrates that the interplay between aerosol loading and cloud
 1692 microphysics is highly sensitive to the prevailing meteorological conditions. While E3SMv2 reliably
 1693 reproduces the overall trends in aerosol effects on stratiform clouds, its performance degrades in deeper,
 1694 more dynamically complex regimes. Given uncertainties in the satellite observations, it is critical for
 1695 future studies to integrate datasets from airborne, ground-based, and satellite platforms. This strategy
 1696 would enable the quantification of errors as well as corroborating the results presented here. In terms of
 1697 the feasibility of potential model improvements, we think that a feasible approach would be the fine-
 1698 tuning of the microphysical parameterization, ideally constrained by high-resolution observational data
 1699 from field campaigns such as ARM. This may reduce the persistent uncertainties in simulating aerosol-
 1700 cloud interactions, particularly under the dynamic meteorological transitions typical of the ENA region.
 1701 Furthermore, emulation from high-resolution modeling (e.g., LES) of cloud and rain microphysics
 1702 processes can be used to replace the bulk microphysics scheme, which can contribute to better
 1703 performance with manageable cost as shown in previous studies such as Gettelman et al. (2021).
 1704 Increasing spatial resolution is also feasible in a regionally refined mesh, and increasing vertical
 1705 resolution might follow, but both would noticeably increase computational cost, so trade-offs should be
 1706 considered with caution. Lastly, the development of new schemes that bridge the gap between shallow
 1707 and deep cloud regimes remains particularly challenging, as current large-scale model schemes still treat
 1708 them separately.

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1720 Future research will focus on exploring the transferability of this regime-based analysis to other
1721 global marine regions, assessing the scaling effects and exploring the process-level understanding of
1722 aerosol-cloud interactions within models, and extending the investigation to include aerosol-cloud-
1723 radiation interactions, thereby providing better constraints on effective radiative forcing. Such efforts are
1724 essential to refine microphysical parameterizations and enhance the overall fidelity of climate models in
1725 representing these critical processes.

Code and Data availability

The E3SMv2 nudged simulation output is available at: <https://zenodo.org/records/15670340>. The ERA5 reanalysis is available at: <https://cds.climate.copernicus.eu/datasets/reanalysis-era5-pressure-levels?tab=overview>. The original clustering model is available at: <https://zenodo.org/records/14720991>. The hyperparameter-tuned model and the collocated CALIPSO-MODIS dataset are available upon request.

Author contributions

The idea of this study was developed by XZ, YF, and DP. XZ performed the analyses and wrote the manuscript under the supervision of YF. MZ performed the nudged E3SM simulation. DP and ZL constructed the collocated CALIPSO-MODIS dataset. XZ, YF, DP, MZ, SX, ZL, RJ and BL participated in further scientific discussions and provided substantial comments and edits on the paper.

Competing interests

At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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